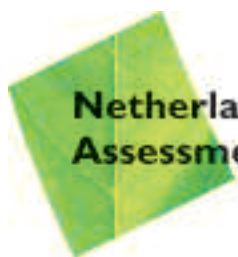


# **Particulate matter: a closer look**

**The state of affairs in the particulate matter dossier from a Dutch perspective**



**Netherlands Environmental  
Assessment Agency**

*Particulate matter: a closer look* is a publication of the Netherlands Environmental Assessment Agency (MNP) in cooperation with the Environment and Safety Division (MEV) of the National Institute for Public Health and the Environment (RIVM).

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## Foreword

With the publication of *Particulate matter: a closer look*, the Netherlands Environmental Assessment Agency and the Environment and Safety Division of the National Institute for Public Health and the Environment want to present the facts about particulate matter in a coherent fashion. This publication summarises the current state of affairs in the particulate matter dossier: what do we know, what don't we know, where are the uncertainties? This publication came about as a result of the current debate in politics and society on the consequences of implementing the Netherlands Air Quality Decree, which is based on directives from the European Union. The limit values for airborne particulate matter are exceeded on a large scale in the Netherlands. The social consequences of these violations are far-reaching; this is because new spatial developments, such as housing construction and infrastructure projects, are liable to be postponed or even cancelled. Moreover, important detrimental health effects are also attributed to airborne particulate matter. The particulate matter dossier is complex and contains administrative dilemmas, legally-binding limit values, concerns of citizens, scientific uncertainties and consequences for spatial planning and the economy. We decided to write a scientific summary report about the particulate matter dossier to answer the many questions that have arisen and to contribute to the current discussions. This publication does not contain any new information, but is a summary of existing reports in the area of particulate matter.

This report addresses six questions:

1. *What is the problem?* The first chapter discusses why there is actually a particulate matter problem. What is particulate matter composed of? How much particulate matter is there in the air? What is the legislation concerning particulate matter? And is this legislation being complied with?
2. *Do other countries also have a problem?* The second chapter presents the measurement data, the instrumentation that is used and the formal reports to the European Commission which make it possible to compare the situation in the Netherlands with the European context.
3. *How high is the emission?* This chapter provides insight into the current and future particulate matter emissions in the Netherlands and a number of neighbouring countries. Which sectors are responsible for the emissions?
4. *How much particulate matter do we measure?* The fourth chapter addresses questions such as: how and where do we measure particulate matter, and how many monitoring stations are there? How is particulate matter measured? What about correction factors? What is particulate matter composed of? And what is the effect of meteorology?
5. *How much particulate matter do we calculate?* This chapter discusses models, which are an important instrument for understanding particulate matter levels. It explains how these models are used and describes the results provided by these models.

6. *What are the health effects?* Particulate matter is given attention primarily due to its detrimental health effects. The final chapter address issues such as: what are these detrimental health effects? How do we know these effects exist? And how certain are we of these effects?

*Particulate matter: a closer look* aims to inform the reader about the particulate matter problem. If you want to explore specific aspects, an extensive reading list is included at the end of the report. The Internet is also a good source of information; links to relevant sites have therefore also been included.

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## Particulate matter considered

### The main conclusions

#### What do we know for certain in the particulate matter dossier?

- Airborne particulate matter can lead to a wide range of detrimental health effects, including premature mortality. It is estimated that several thousand people die in the Netherlands annually in connection with short-term exposure to particulate matter. The reduction in life expectancy is very small.
- To reduce these health risks, the European Union established air quality norms in the form of limit values. All Member States have had to comply with these limit values since 1 January 2005. The limit values also apply to areas where no people live.
- Particulate matter concentrations are measured in the Netherlands according to the method prescribed by European legislation. The measurement and modelling instruments that are used have a level of reliability that meets the requirements in the relevant European legislation.
- Between 1992 and 2003, the concentration of airborne particulate matter declined by  $1 \mu\text{g}/\text{m}^3$  per year on average. The total decline in particulate matter concentration since 1994 has been 25%.
- Between 1990 and 2003, the emissions in the Netherlands from known sources of particulate matter and gases from which particulate matter can be formed in the air have declined sharply. This is because many measures were taken during this period in the Netherlands, such as switching from oil to natural gas.
- Measurements and model calculations show that the limit value for the annual average concentration ( $40 \mu\text{g}/\text{m}^3$ ) is exceeded in the Netherlands, but only to a limited extent.
- The limit value for the 24-hour average concentration (no more than 35 days per year exceeding a 24-hour average concentration of  $50 \mu\text{g}/\text{m}^3$ ) is exceeded in large areas of the Netherlands.
- Violations of the limit values have been observed in nearly all European cities. The violations in the Netherlands, Belgium, Germany and Italy take place over a larger geographical area than in other Member States.
- At least 45% of the average particulate matter concentration in the Netherlands is of anthropogenic origin. The other 55% originates primarily from sea salt, soil dust and unknown or incorrectly estimated sources.

- It is estimated that two-thirds of the anthropogenic particulate matter originates from sources outside the Netherlands and that one-third originates from within the Netherlands. However, due primarily to the effect of local traffic, on busy streets the concentration originating from within the Netherlands can rise to 30-45%.
- Despite the high contribution from other countries, the Netherlands is still a net exporter of particulate matter. The Dutch 'export' of particulate matter is three times as large as the 'import'.

## **What are the uncertainties in the particulate matter dossier?**

- Not all detrimental health effects are known. There are indications that, in addition to short-term exposure, it is especially long-term exposure to particulate matter that causes detrimental health effects. Estimates vary from possibly ten thousand to several tens of thousands of people who die approximately ten years prematurely. This amplifies the relevance of the current limit values.
- The European air quality directives allow for multiple administrative and technical interpretations. This leads to differing implementations in the Member States; as a result, in Europe there is no level playing field regarding the protection of public health.
- The modelling method calculates non-compliance with a maximum uncertainty margin of 50%. As a result, the amount the limit values are exceeded also has an uncertainty margin. These uncertainties are not considered in the judicial analysis. The average estimate is used to determine compliance with the limit values, and measurements and model results are used as if they were absolute values.
- In view of the high level of uncertainty in determining particulate matter concentrations, there is a risk that building projects will be suspended in areas where the estimated concentration lies just above the limit value, and the actual concentration lies just below the same limit value. The other way around, there is a risk that projects will be continued at locations where the estimated concentration lies just below the limit value, but the actual concentration is just above the same value. Such risks are inherent to environmental problems where concentrations fluctuate around the limit value.
- The Netherlands is currently in non-compliance with the limit values of the European Union, and this situation is expected to continue for the near future. It can be expected that the European Commission will require the Netherlands to take all policy measures that are within reason to solve this problem. It is still unclear what 'within reason' entails.



## How do we proceed?

- Due to further reductions of particulate matter emissions in the Netherlands and especially in neighbouring countries, the air quality in the Netherlands will continue to improve. Nevertheless, the limit value for the 24-hour average concentrations along highways and in inner cities is expected to be exceeded for a number of years to come. To comply with the limit values, the uncertainties in the particulate matter dossier are not leading at the present time to policy measures that would be regretted afterwards. Since detrimental health effects still exist when the concentration falls below the limit value, public health is benefited by every measure to reduce particulate matter concentrations.
- The policy in the Netherlands is based on the combination of measurements and calculation models with the aim of achieving the best possible picture of reality. In many other countries, interpretations based only on measurements are thought to be sufficient. The downside of this approach is that it underestimates the actual situation. What's more, it is impossible to evaluate future situations based solely on measurements.
- The current limit values do not make a distinction between the various fractions of particulate matter. All fractions are treated as if they were equally relevant to health. By disregarding non-hazardous particulate matter fractions of natural origin, such as sea salt, it is easier to comply with limit values and spatial planning limitations can be partly eliminated. However, this does not reduce the health risks of particulate matter.
- The particulate matter problem cannot be solved by the Netherlands alone. A European-wide approach is required. Supplemental European source policy – focusing primarily on reducing traffic emissions – is cost-effective for the Netherlands. Such a policy reduces both domestic pollution and the import of pollution from abroad. To comply with the limit values, the Netherlands will also have to take supplementary measures. This is because the Netherlands is a densely populated country with a great deal of industry and transport.
- Although it is still unclear which particulate matter fractions are most relevant to health, there are indications that traffic emissions play an important role. A policy that focuses on the soot fraction of particulate matter is sensible from a health point of view and appears to be most probably a 'no regret' approach. However, other components in traffic emissions must also be considered in this context.



## 1. What is the problem?

- Public health studies indicate that in the Netherlands, several thousand people die prematurely each year related to short-term exposure to particulate matter. The duration of this reduced life expectancy is probably very short, ranging from several days to months. Similar results have not only been found in the Netherlands, but everywhere in the world, and these results are fairly robust.
- If certain American studies concerning long-term exposure are applied to the Netherlands, it is possible that ten thousand to several tens of thousands of people die approximately ten years prematurely. However, these results are extremely uncertain.
- Air quality in terms of particulate matter has improved in the Netherlands during the past ten years. The concentration of particulate matter has declined by 25%. Nevertheless, the European limit values are still exceeded in the Netherlands. This will also be true in the near future.
- Non-compliance with the air quality limit values appears to be a reason to hold back planned spatial developments.
- At least 45% of the particulate matter components are of anthropogenic origin and at least 15% originate from sources in the Netherlands. In urban areas, the anthropogenic contribution from sources in the Netherlands is 30-45%, especially due to traffic.

### *Overview of the chapter*

The first chapter briefly describes the most important aspects of the problems surrounding particulate matter. The questions addressed are: what is particulate matter, what are its components, what are its detrimental health effects and what are the concentrations of particulate matter in the Netherlands? The chapter also addresses European legislation, the Dutch framework, the policy context and administrative complications. In the following chapters, specific aspects of the particulate matter dossier will be examined more deeply.

### **Problem statement**

In 1999, the European Union established two air quality norms for particulate matter: a limit value for the annual average concentration and a limit value for the 24-hour average concentration (EU, 1999). Internationally-accepted insights about the detrimental health effects of particulate matter are contained in this legislation (WHO, 2000). The limit values apply Europe-wide and have been implemented in Dutch legislation (Staatsblad, 2001). Testing to determine compliance with the limit values takes place, among other ways, by measuring particulate matter concentrations. These measurements take place using a method prescribed by the European Union. These measurements show that the limit value for annual average annual particulate matter concentrations is exceeded by a limited amount. The limit value for 24-hour average concentrations, in contrast, is exceeded on a large scale. This will probably continue to be the case in the future.

The majority of the particulate matter concentrations cannot be influenced by Dutch policy. The particulate matter problem is therefore very recalcitrant, and for the Netherlands alone it is virtually insoluble. Nevertheless, the European Commission requires the Netherlands to make every reasonable effort to comply with the limit values. Densely-populated regions and countries such as the Netherlands are confronted with the consequences of uniform air quality norms to guarantee their citizens at least a minimum level of health protection. Compared to other countries, this leads to extra costs for Dutch society due to limitations placed on spatial development or the necessity to take supplementary policy measures.

There are major scientific uncertainties in the particulate matter dossier; these uncertainties concern the emissions, the measurements, the models and the detrimental health effects. In view of these uncertainties, the Dutch Cabinet is faced with the challenge of choosing measures that are the most robust, that provide the most health benefits and that are the most cost effective. Moreover, the measures must be applicable both legally and administratively, and they must have social support.

## Particulate matter

Particulate matter is a type of air pollution in particle form. Particulate matter is a complex mixture of particles of various diameters and various chemical compositions. A widely-used abbreviation for particulate matter is PM. Depending on the diameter of the particles, either the abbreviation PM<sub>10</sub> is used (for particles with a diameter up to 10 micrometers) or the abbreviation PM<sub>2.5</sub> (for particles with a diameter up to 2.5 micrometers)<sup>1</sup>. In the remainder of this publication, when the term particulate matter is used, it will refer to PM<sub>10</sub>. If the term particulate matter is used with a different meaning than PM<sub>10</sub>, then this will be expressly stated.

## Components

In chemical terms, particulate matter is not a simple and unambiguous concept. Important components of particulate matter include soil dust, sea salt and anthropogenic emissions (caused by human activities). The latter component concerns substances from direct emissions, the so-called primary emissions, and substances that have been created by chemical reactions in the atmosphere, such as sulphur dioxide (SO<sub>2</sub>), nitrogen oxides (NO<sub>x</sub>) and ammonia (NH<sub>3</sub>), the so-called secondary aerosol. In addition, other substances can be present in smaller amounts, but which are still relevant to health.

It is possible to make a further distinction according to the size of the particulate matter. The fraction PM<sub>2.5</sub> contains the fine and ultra-fine particles. These are primarily the particles originating from the condensation of combustion products or the

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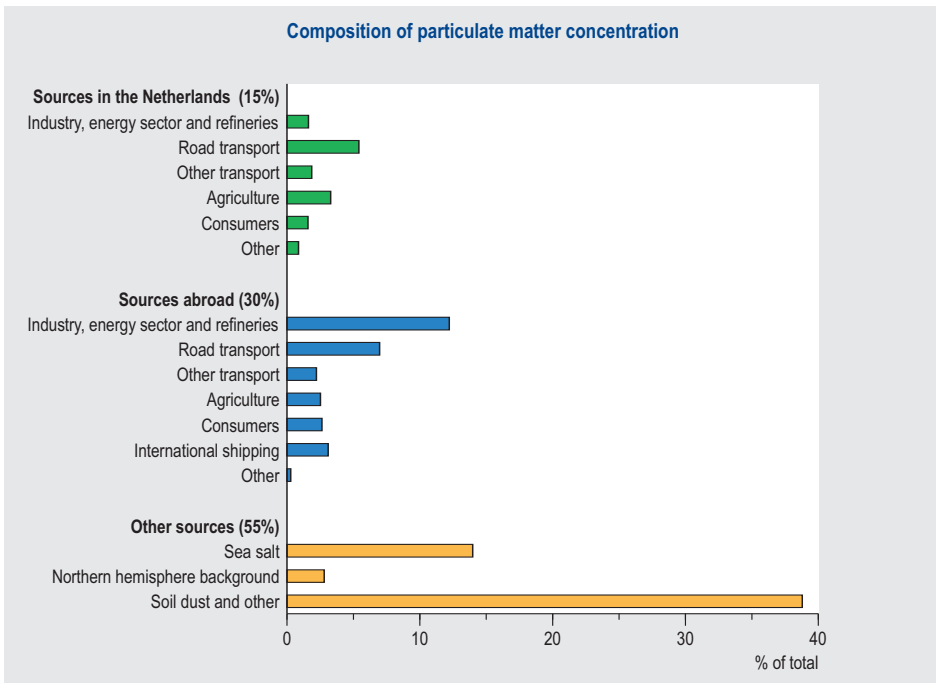
<sup>1</sup> PM<sub>10</sub> and PM<sub>2.5</sub> are good approximations of the mass of particles with diameters up to 10 µm or 2.5 µm, respectively.

reaction of gaseous pollutants. The fraction larger than  $PM_{2.5}$ , indicated with the abbreviation  $PM_{2.5-10}$ , comprises primarily mechanically-formed particles. Anthropogenic contributions to this fraction primarily originate from windblown traffic-related dust, such as dust caused by tyre wear, and dust emissions from animal husbandry. Chapter 3, *How large high is the emission?*, provides more information about emissions and future developments of emissions.

The composite particles of particulate matter, depending on their size, have an atmospheric residence time ranging from days to weeks. As a result, particulate matter can move over distances of thousands of kilometres; it is therefore a problem at the continental scale.

## Origin

According to model calculations, at least 45% of particulate matter components are of anthropogenic origin. Of this fraction, two-thirds originates outside the Netherlands and one-third from inside the country (*Figure 1.1*). From this it follows that at least 15% of the total particulate matter concentrations can be influenced by Dutch policy. The



*Figure 1.1 Average composition of particulate matter concentrations in non-urban areas in the Netherlands subdivided according to source contributions. ‘Soil dust and other’ in the category ‘Other sources’ is the many-year average of the non-modelled portion of particulate matter comprising biological matter, water and the contribution from sources that are not modelled or have been incorrectly modelled. As a result, this may partly include anthropogenic sources. For a more complete explanation, see Chapter 4, ‘How much particulate matter do we measure?’, and the text box, ‘Chemical composition of particulate matter in the Netherlands’.*

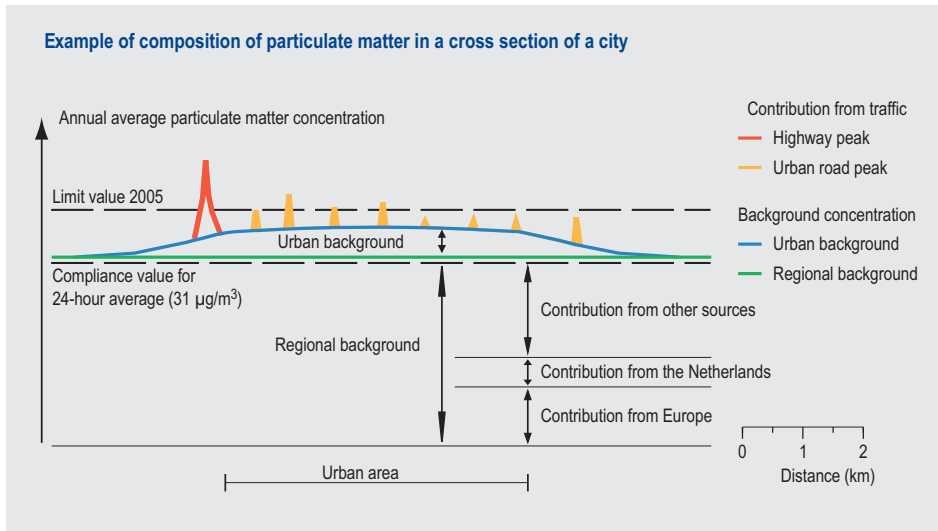


Figure 1.2 Composition of particulate matter concentrations in an urban area. Source: MNP, 2005. 'Contribution from other sources' is the many-year average of non-modelled particulate matter. This is composed of sea salt, the northern hemisphere background, soil dust, biological matter, water and the contribution of non-modelled or incorrectly-modelled sources. For a more extensive explanation, see Chapter 4, 'How much particulate matter do we measure?', and the text box, 'Chemical composition of particulate matter in the Netherlands'. See also Figure 1.1. The uppermost horizontal dotted line indicates the limit value for the annual average concentration,  $40 \mu\text{g}/\text{m}^3$ . The lowermost horizontal dotted line is equivalent to an annual average concentration of  $31 \mu\text{g}/\text{m}^3$ . This is the annual average concentration where the limit value for the 24-hour average is not exceeded. For a further explanation of the above, see Figure 4.3 in Chapter 4, 'How much particulate matter do we measure?'

other 55% is composed largely of contributions from sea salt, soil dust, the large-scale northern hemisphere background and unknown and possibly incorrectly-modelled anthropogenic sources (Visser *et al.*, 2001).

In urban areas along streets, the national anthropogenic contribution can rise to 45% of the total concentration. This is primarily caused by the local traffic (Figure 1.2). More information about the composition and origin of particulate matter concentrations is presented in Chapter 5, *How much particulate matter do we calculate?*

## Legislation

During the second half of the 1990s, the detrimental health effects of particulate matter resulted in legislation being passed in the European Union. The European legislation contains limit values for particulate matter concentrations. These limit values are interim goals; the ultimate aim is to achieve sustainable levels (EU, 1996). Two limit values for particulate matter have been defined. Both aim to protect human health. The first limit value for particulate matter concerns the annual average concentra-

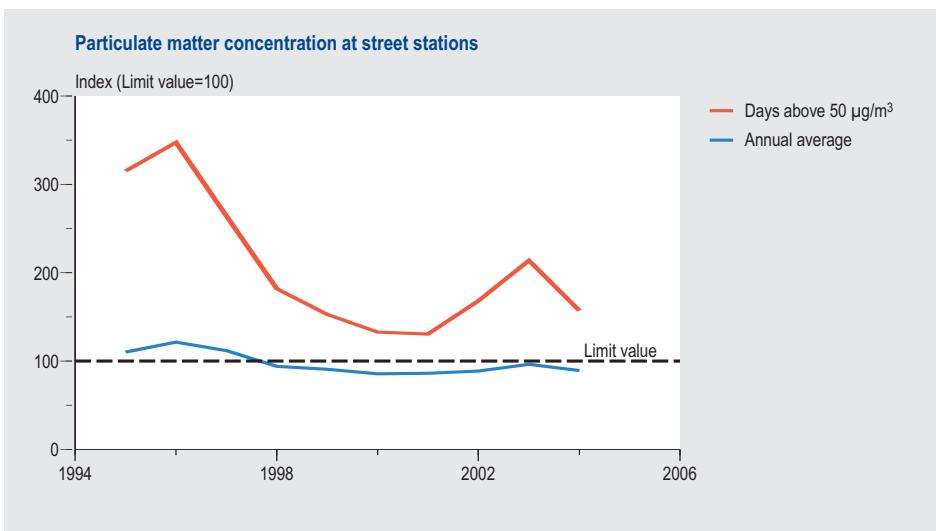
tion. This value must not exceed  $40 \mu\text{g}/\text{m}^3$ . The second limit value is the 24-hour average concentration. Exceeding a 24-hour average particulate matter concentration of  $50 \mu\text{g}/\text{m}^3$  is not allowed for more than 35 days per year. All Member States have been required to comply with both limit values since 1 January 2005. A further explanation of the legislative aspects is given in the text box, *Legislation*.

## Air quality

During the past ten years, the air quality in terms of particulate matter has improved in the Netherlands (*Figure 1.3*). The annual average concentration declined during this period by 25%. In fact, the number of days with a 24-hour average concentration above  $50 \mu\text{g}/\text{m}^3$  declined by a factor of two. Nevertheless, both limit values are still being exceeded in the Netherlands. It appears that the limit value for the 24-hour average is exceeded on a larger scale than the value for the annual average concentration (*Figure 1.4*).

For that matter, the Netherlands is not the only European country that does not comply with the limit values. The urban air quality in the Netherlands is similar to that in other European countries. These aspects will be discussed more extensively in Chapter 2, *Do other countries also have a problem?*

Based on the current policy it is expected that there will be nearly full compliance with the limit value for the annual average concentration in 2010 and full compliance



*Figure 1.3 Development of the air quality for particulate matter at street stations. Since 1995, the air quality for particulate matter has clearly improved. The lines indicate the average development in the Netherlands based on the measurement results at the street stations. In recent years, the limit value for the annual average concentration (blue line) in the Netherlands has been exceeded at only a few locations. In contrast, the limit value for the 24-hour average concentration is still being exceeded on a large scale (red line). Source: MNP, 2005.*

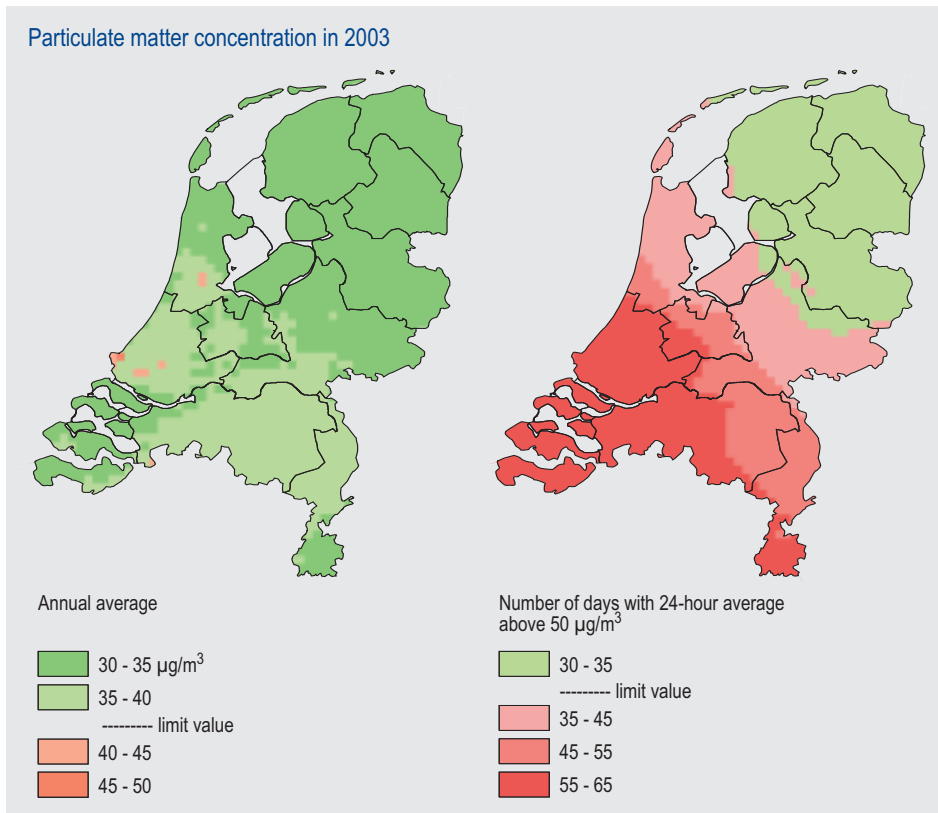


Figure 1.4 Annual average particulate matter concentrations (left) and the number of days with a 24-hour average particulate matter concentration above 50  $\mu\text{g}/\text{m}^3$  (right) in the Netherlands in 2003, shown on a grid with  $5 \times 5$  km cells. The limit value for the annual average concentration is still being exceeded in the Amsterdam, The Hague and Rotterdam regions, but on a very limited scale. In contrast, the limit value for the 24-hour average is exceeded in more than half of the country.

The map for the annual average concentrations was obtained from measurement results originating from the Dutch National Air Quality Monitoring Network combined with model calculations. The map for the number of days exceeding the limit value was constructed by interpolation of the measurement results from the regional monitoring stations in the Dutch National Air Quality Monitoring Network. Source: MNC, 2005.

in 2020. Although improved compliance with the limit value for the 24-hour average is expected, it is likely that this limit value will still be exceeded in 2020, especially in cities and in the vicinity of highways (Folkert *et al.*, 2005). The air quality in the Netherlands is presented in more detail in Chapter 4, *How much particulate matter do we measure?*

## Health effects

Particles smaller than ten micrometers in diameter enter the tracheobronchial airways during inhalation. As a result, particulate matter in the air can lead to health



## Legislation

In 1996, the Air Quality Framework Directive went into force (EU, 1996). The Framework Directive provides a new and coherent general European framework for 'evaluating and managing air quality'. The Framework Directive uses a number of important concepts: daughter directives, preliminary assessments, assessment thresholds and zones and agglomerations. The daughter directives are specifications of air quality requirements for certain substances. In the meantime, four daughter directives have appeared (EU, 1999; EU, 2000; EU, 2002; EU, 2005).

The concentration levels of substances from the first daughter directive, including particulate matter, have been an important element in the definition of the zones and agglomerations in the Netherlands (Van Breugel and Buijsman, 2001). The result has been a subdivision of the Netherlands into three zones and six agglomerations (Figure 1.5). The agglomerations are urban areas with at least 250,000 residents. Moreover, the first daughter directive stipulates the numbers of monitoring stations in the zones and agglomerations, which are in turn dependent on the numbers of residents and the concentration levels. The Directive also contains regulations concerning the monitoring apparatus to be used. The implementation of these aspects in the Netherlands has taken place entirely in accordance with the European Directive. This was included in

Dutch legislation in 2001 as part of the Air Quality Decree (Staatsblad, 2001).

The Directive stipulates two limit values. There is a limit value for the annual average concentration of particulate matter that is primarily intended to offer protection against the long-term effects of particulate matter. This limit value is  $40 \mu\text{g}/\text{m}^3$ . The second limit value concerns the 24-hour concentration of particulate matter. This is primarily intended to provide protection against the short-term effects. Specifically, the Directive stipulates that the limit value for the 24-hour average ( $50 \mu\text{g}/\text{m}^3$ ) cannot be exceeded for more than 35 days during each calendar year.

The Directive originally assumed that the two limit values were equivalent; based on the knowledge at that time they were they thought to be equally 'stringent'. In practice, this has turned out not to be the case. The limit value for the 24-hour average is more 'stringent' than that for the annual average concentration. This topic is discussed in more detail in Chapter 4, *How much particulate matter do we measure?*

The European legislation offers possibilities to subtract particulate matter originating from 'natural phenomena' from the measured particulate matter concentrations under certain conditions (EU, 2001).

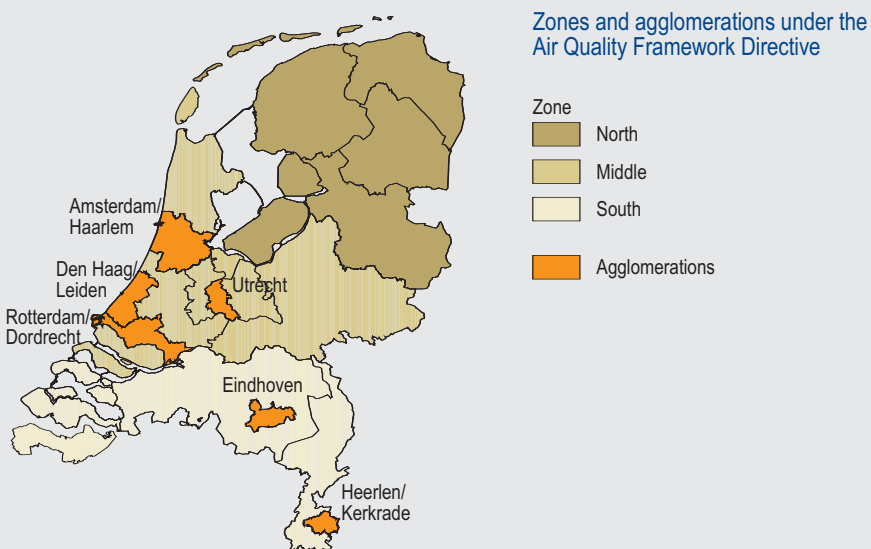


Figure 1.5 The division of the Netherlands into zones and agglomerations in accordance with the Air Quality Framework Directive (Van Breugel and Buijsman, 2001).

problems and even to premature mortality. Epidemiological studies indicate that 2300 to 3500 people die prematurely every year in the Netherlands due specifically to the acute consequences of exposure to particulate matter. Based on the long-term effects of chronic exposure to particulate matter, it is possible that as many as 12,000 to 24,000 people die prematurely from this cause every year in the Netherlands.

Moreover, research has shown that there is probably no threshold value below which no detrimental health effects occur. A complicating factor is that it is not well understood which components of particulate matter are most responsible for these effects. From this it follows that a reduction of the emissions that contribute to particulate matter concentrations could lead to a reduction of the concentrations, but this will not necessarily lead to a reduction in the magnitude of the detrimental health effects. Consequently, there are two partly-related problems: it is one problem to meet the requirements of the legislation and a second problem to reduce the detrimental health effects. The detrimental health effects will be discussed more extensively in Chapter 6, *What are the health effects?*

## Problems in the Netherlands

Based on the particulate matter levels measured in 2002 in the Netherlands, the country was required to develop plans for supplementary measures to improve the air quality. The Dutch Cabinet met this obligation by passing the National Air Quality Plan 2004 (NPL04). The aim of the plan is to indicate which supplementary measures should be taken to comply with the limit values for particulate matter within the established deadlines. In the Netherlands, European emission requirements resulted in a sharp decline in emissions from traffic and industry. However, due to the dense population and building density in the Netherlands, this was not enough to meet the European environmental requirements (Beck *et al.*, 2005a). The problems in the Netherlands are also related to the fact that much of the pollution in the country originates from abroad. The text box *What is the Netherlands doing?* provides more information about Dutch policy.

To comply with the European limit values for particulate matter, extra measures are therefore required. During this process, tension can develop between the competitive position of the Netherlands and the European aim for equal protection of its citizens against excessive air pollution. Moreover, the Netherlands has implemented the EU Air Quality Directive to the letter, which means that the Netherlands is dealing more stringently with the limit values than other European countries.

## Administrative-judicial aspects

Since the Air Quality Decree went into force in 2001, a judicial regime has gone into effect where construction and expansion plans can be blocked or modifications to the plans can be required. This is shown from decisions of the Litigation Section of the Council of State (the highest judicial authority in the Netherlands). In the meantime,

**PM<sub>2.5</sub>**

In mid-2005, the European Commission will present a strategy to continue to deal with the negative effects of air pollution on people and the environment. This is taking place in the context of the Clear Air for Europe (CAFE) programme. In support of this programme, the World Health Organisation recommended in a recent evaluation of the detrimental health aspects of air pollution that PM<sub>2.5</sub> be used as an indicator. It believes that this fraction has a greater impact on human health than PM<sub>10</sub>. This recommendation led the European Commission to propose legislation concerning PM<sub>2.5</sub>. The PM<sub>2.5</sub> fraction is linked more directly with the anthropogenic emission of particulate matter and can therefore be more successfully controlled with policy measures.

Components of natural origin, such as sea salt and some soil dust, play a much smaller role in the PM<sub>2.5</sub> fraction than in the PM<sub>10</sub> fraction. However, there are also practical disadvantages to

possible legislation concerning PM<sub>2.5</sub>. Measurements of PM<sub>2.5</sub> are taking place in Europe only on a limited scale. It is estimated that there were only 90 monitoring stations in Europe in 2003 (AIRBASE, 2005). At the time this report was completed, the Netherlands had only three regional monitoring stations and two street stations for measuring PM<sub>2.5</sub>. The measured annual average concentration of PM<sub>2.5</sub> is approximately 15–25 µg/m<sup>3</sup>. The differences in concentration between busy roads, the urban background and the rural area appear to be small. However, there are still too few measurement results available to provide a good picture of this situation in the Netherlands. There is also little reliable data about the magnitude of PM<sub>2.5</sub> emissions and of the effect of policy measures on these emissions. It is expected that the European Commission will make agreements with the Member States for 2005/2006 about the limit value for PM<sub>2.5</sub>. The agreements will also involve emission ceilings for PM<sub>2.5</sub> on a country-by-country basis.

more than 40 objections to spatial development plans have been lodged with various judicial authorities, including the Council of State, due to possible conflicts with the Air Quality Decree. In one-third of these cases, the Council of State nullified a plan based on the Air Quality Decree. This concerns, for example, zoning plans for residential construction or industrial developments, permits for new business activities and plans for building or modifying roads or highways. The decisions of the Council of State make it clear that before such plans are approved, a very careful analysis must be conducted into the consequences for air quality. Failing to comply with the air quality limit values can be a reason for holding back spatial developments. Moreover, it is possible that the decisions of the Council of State do not reflect the entire problem. A recent initial survey of the Association of Netherlands Municipalities showed that around half of the municipalities in the Netherlands have problems, or believe they will have problems, with the consequences of air quality norms on spatial planning issues (VNG, 2005). According to this survey, plans for more than 100,000 residences and 4500 hectares of industrial developments are faced with postponement or cancellation.

## European developments

An evaluation of the European limit values for particulate matter is part of the Clean Air for Europe (CAFE) programme. This is a programme of the European Commission to improve the air quality in the European Union to a level where ‘there are no longer any significant negative effects’ on human health or the environment. Particulate matter is included in this programme. Two new aspects are the attention to the finer fraction of particulate matter, PM<sub>2.5</sub>, and the discussion about the possibility of making a statutory exception for components in particulate matter that are of natural

origin and which are not viewed as hazardous. An example of such a component is sea-salt aerosol. The current limit values for particulate matter (annual average and 24-hour average) will be continued.

### **What is the Netherlands doing?**

The policy of the Dutch Cabinet in the particulate matter dossier focuses on two points: reducing health risks and reducing the risks that new spatial developments will stagnate. The Cabinet is taking three tracks to solve the air quality bottlenecks:

#### **Application of national measures that improve air quality**

The accent here is on a series of subsidy and stimulus measures that accelerate and increase the implementation level of soot filters on diesel cars and trucks in the Netherlands. In addition, the purchase of clean Euro-4/5 trucks and Euro-5 diesel automobiles is being stimulated with tax measures. The maximum speed on highways is being reduced to 80 kilometres per hour at five highway routes that are particulate matter 'hotspots'. The Cabinet plans to supplement this package of measures with several budget-neutral measures and with local measures implemented by provinces and municipalities. The package of measures will be presented in the autumn of 2005 and will be completed with the National Air Quality Plan (NLP05) that will appear at the end of 2005.

#### **Evaluation and modification of air quality norms in a European context**

The NLP05 will be used in Brussels to show that the Netherlands is making every effort that can be reasonably expected from the country. The Cabinet is expecting the European Commission to be accommodating, which will reduce the risk of the Netherlands being declared in non-compliance with the Air Quality Directive.

At the end of 2005, the European Commission will publish a thematic strategy which will map out the contours of future European air quality policy, therefore including Dutch policy. In this context, the Netherlands has argued in favour of consistency between the emission and air quality policy

in the European Union and for a more stringent approach to the emission reduction policy for specific sources. In addition, the Netherlands has made proposals to focus particulate matter policy on hazardous combustion emissions. The extent to which this strategy has been successful and can lead to a solution of the current bottlenecks will become clear as soon as the thematic strategy appears.

#### **Clarification of Dutch legislation**

The decisions of and the advice provided by the Council of State have resulted in the Air Quality Decree being amended. New elements in the Decree include the following: a debit/credit approach for construction plans in situations where the limit value is exceeded; the non-hazardous natural component of particulate matter will be disregarded; and the 'stand still' principle in the Environmental Protection Act will be abolished. The credit/debit approach, combined with abolishing the stand still principle, will be worked out in detail in a Ministerial order and should result in jurisprudence. This approach will possibly result in a shift of emphasis from individual building and development projects to the developments in air quality for an entire zone or agglomeration. If there is a positive recommendation from the Council of State, the amended Air Quality Decree will go into force in the Summer of 2005. The new Air Quality Decree will then be replaced by an Act. The Cabinet will present a proposal for this in the Autumn of 2005.

*As this publication went to press, there was still a great deal of movement concerning the three points mentioned above. It goes without saying that the effects of Cabinet policy on air quality can only be evaluated when the package of measures has been finalised and has also been sufficiently instrumentalised. The MNP will present an evaluation of the effectiveness and efficiency of the Cabinet's proposals in September 2005.*

**Particulate matter and climate**

Particulate matter also plays a role in the enhanced greenhouse effect (IPCC, 2001). Particulate matter is usually indicated in this context with the term 'aerosols'. Aerosols can absorb sunlight falling on the earth, but they can also reflect it. Which of these behaviours an aerosol displays depends on its chemical composition. The majority of components, such as sulphur and nitrogen aerosols and organic carbon, reflect sunlight and therefore have a cooling effect. Soot absorbs sunlight and therefore has a heating effect. This suggests that an approach that primarily focuses on soot could be beneficial. It has both a positive health effect and a climatic effect by partly neutralising the enhanced greenhouse effect, especially at the regional scale. A decline in the concentration of other aerosols, however, would lead to an increase of the enhanced greenhouse effect.

Aerosols and carbon dioxide frequently originate from the same sources. For example, they are

simultaneously emitted during combustion processes. To calculate the total effect of source measures on climate, both products must be taken into account. One example is traffic. Modern diesel autos are 20% to 30% more efficient than comparable petrol autos and therefore emit 10% to 20% less carbon dioxide for each kilometre travelled. This is beneficial for counteracting the enhanced greenhouse effect. On the other side of the equation is the higher emission of soot particles by diesel autos in comparison with petrol autos. This has a warming effect on the climate at the local scale, which cannot yet be properly quantified. In addition, the fuel costs of a diesel auto are 40% to 50% lower than those of a comparable petrol auto. As a result, drivers who switch from a petrol auto to a diesel auto tend to drive more. In this way, a portion of the reduction of carbon dioxide emissions is counteracted. The net effect depends, among other things, on the implementation degree of soot filters on diesel autos.



## 2. Do other countries also have a problem?

- The Netherlands is certainly not the only country that has a problem. In nearly all urban areas in Europe there have been reports of the European limit values for particulate matter being exceeded. The levels of particulate matter in such areas are comparable with those in the Netherlands.
- However, the limit values are exceeded on a larger scale in the Netherlands. This situation is comparable with urban areas such as those in Belgium, the Ruhr district in Germany and the industrialised area of Northern Italy.
- There are important differences between countries in their implementation of the relevant European legislation. For example, the Netherlands has implemented the legislation to the letter.
- The Netherlands is one of the few countries to use a set of modelling instruments with a high spatial resolution capacity for analysing and reporting on air quality.

### *Overview of the chapter*

This chapter compares the situation in the Netherlands with that in a number of other European countries. To this end, European measurement data and the formal reports of non-compliance to the European Commission have been used. There is also a brief discussion about the implementation and adaptation of European legislation in other countries.

### **A large-scale problem**

An initial analysis shows that the Netherlands is not the only country where increased particulate matter concentrations occur. The distribution of particulate matter is a large-scale phenomenon. This is shown, for example, from the situation in Germany, where the limit value for the 24-hour average is exceeded in a large number of urban areas (*Figure 2.1*). At monitoring stations in Dortmund, Düsseldorf, Dresden, Hannover, Leipzig and Munich, the maximum number of days allowed for the entire year with a 24-hour average above  $50 \mu\text{g}/\text{m}^3$  had already been exceeded on 1 June 2005 (UBA, 2005a).

### **Measurement data**

One source of information about air quality for particulate matter in the countries of the European Union is AIRBASE, the database with air quality data from the European Topic Centre on Air and Climate Change (ETC/ACC) of the European Environment Agency (EEA). This data also shows that both limit values are being exceeded on a large scale, although here as well the limit value for the 24-hour average is exceeded significantly more often (*Figure 2.2*). Moreover, the data in AIRBASE show that most of the excessive values are concentrated in the measurements from urban stations. These are a good example of monitoring stations that are strongly affected by local sources. A similar picture emerges from the mandatory annual reporting from the

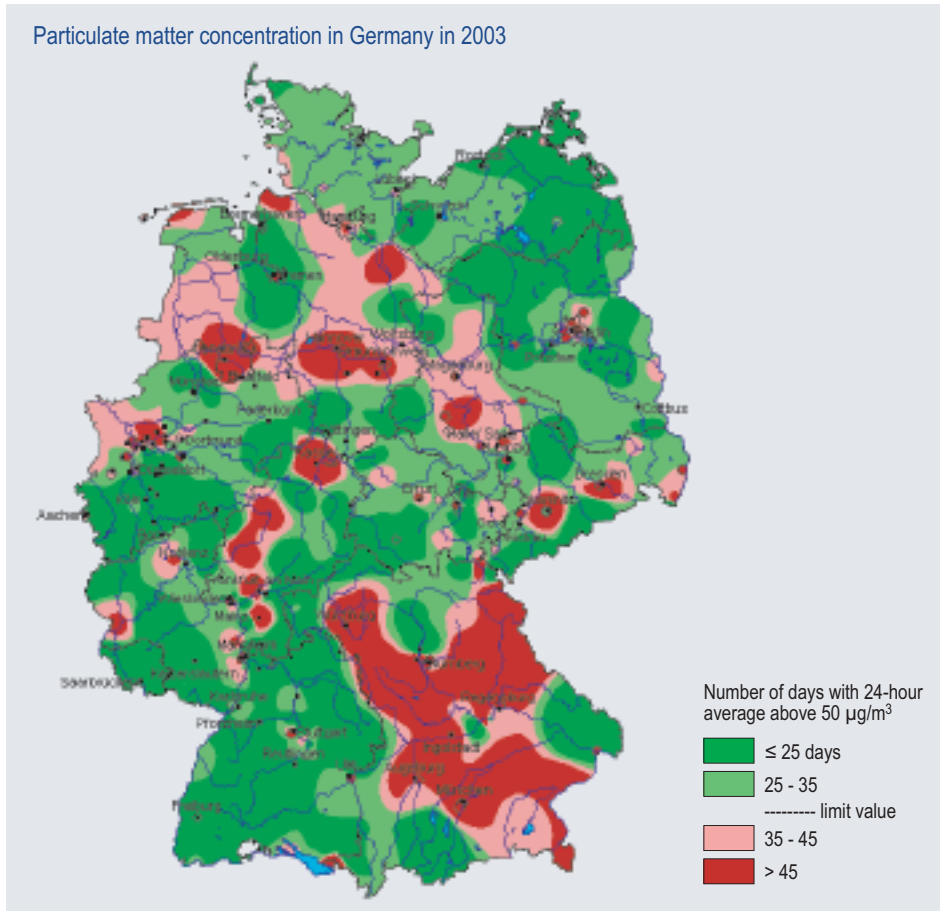


Figure 2.1 Number of days in Germany in 2003 that the 24-hour particulate matter concentration was higher than 50  $\mu\text{g}/\text{m}^3$ . Source: UBA, 2005b.

Member States of the European Union to the European Commission. In most zones and agglomerations in Belgium, Germany, the Netherlands and the United Kingdom, the limit values are exceeded. This picture stands in contrast with the situation in France and other countries, where the limit values are exceeded much less often (Table 2.1). However, it is impossible to draw far-reaching conclusions from this information without involving the exact situation and size of the zones in the various countries. It is possible that some of the differences can be explained by differences in the correction factors used.

### Correction factors

A complicating factor in the use of measurement data for particulate matter was formulated by the CAFE working group as follows: 'Due to differences in calibration of the continuous monitors in relation to the reference method, and due to differences



### Particulate matter concentration in Europe in 2002

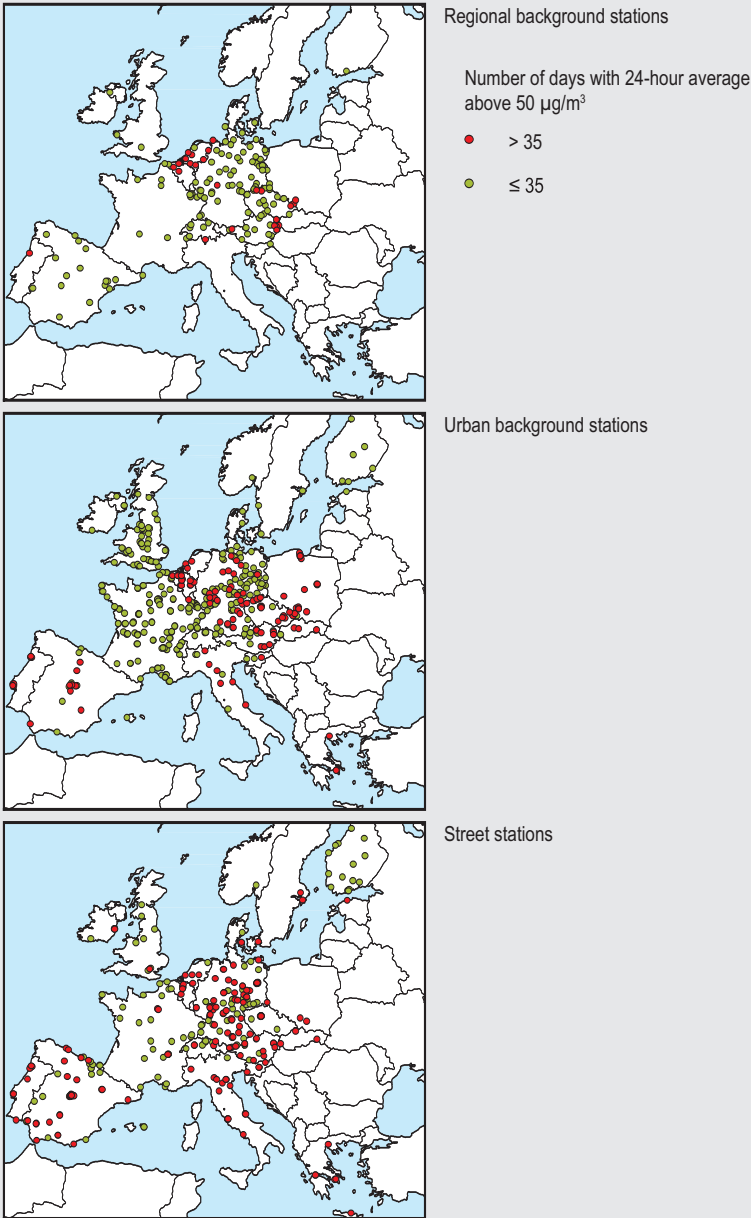


Figure 2.2 Occurrences where the limit value of the 24-hour average concentration of particulate matter in Europe was exceeded in 2002. Source: AIRBASE. Limit value: there must not be more than 35 days per year with a 24-hour average concentration above  $50 \mu\text{g}/\text{m}^3$ . This limit value has applied to all Member States since 1 January 2005. Data from AIRBASE show that the limit value for the 24-hour average is exceeded at 52% of the street stations, at 28% of the urban background stations and at 18% of the regional background stations.

*Table 2.1 Instances where the limit values were exceeded in the zones and agglomerations of a number of European countries in 2003 according to the official reports of Member States submitted to the European Commission. Source: CIRCA, 2005; AirBase, 2005.*

	Total number of monitoring stations <sup>1</sup>	Non-compliance with the limit value for the 24-hour average		Non-compliance with the limit value for the annual average	
		yes	no	yes	no
<i>number of zones and agglomerations</i>					
Belgium	33	10	0	9	1
Denmark	8	2	4	1	5
Germany	367	49	29	13	65
France	232	12	56	5	63
<b>The Netherlands</b>	<b>33</b>	<b>9</b>	<b>0</b>	<b>3</b>	<b>6</b>
Austria	95	11	0	3	8
United Kingdom	72	32	10	14	28

Note: The data for the Netherlands deviate from those presented in Chapter 4, *How much particulate matter do we measure?* because it involves different years.

in the “station mix” in the networks of the Member States, full comparability of PM<sub>10</sub> levels over Europe is not ensured’ (EU, 2004). In fact this means that reported monitoring data cannot be simply compared to each other directly. Not only are there differing monitoring systems, most of the instruments used also make a systematic error (see Chapter 4, *How much particulate matter do we measure?*). This is caused, among other things, by the evaporation of semi-volatile particles during sampling. Based on relevant research results, the Netherlands therefore increases the measured results of particulate matter monitoring by a factor of 1.33. A number of other countries follow the indication provided by the European Commission and use a factor of 1.3. Only Belgium uses higher correction factors. Most countries use a lower correction factor or no correction factor at all. This is usually based on their own research, although for a number of countries it is unclear what the basis is for the value of the correction factor (Buijsman and De Leeuw, 2004).

## Use of models

Besides measurements, countries are also allowed to use models to determine the air quality or ascertain air quality ‘hotspots’. A recent survey (Koelemeijer *et al.*, 2005) showed that only a few countries calculate air quality down to the street level for their reporting to the European Commission; these countries are Denmark, the Netherlands, the United Kingdom and Sweden. Since the measurements show that the limit value is exceeded most frequently at the street level (see *Table 2.1*) the number of hotspots in the countries that do not model their air quality down to the street level – which is the majority of countries – are possibly underestimated.

## Implementation of European legislation

A study was recently conducted into the way in which the various European countries deal judicially with air quality legislation (Bakker, 2004; Backes and Van Nieuwerburgh, 2005; Koelemeijer *et al.*, 2005). It appears that there are major differences between countries. In the Netherlands there is an explicit statutory link between air quality policy and other types of policy, including spatial planning policy. In other countries there is usually not such an explicit link; in various countries only plans with potentially far-reaching effects are subjected to a review. In addition, the Netherlands strictly enforces the limit values. This means that when granting permits, a clear differentiation is made between plans that do not comply with the limit values and those that do comply, even if the relevant plans lead to values that are just below or just above the limit.

In some other countries, implementation takes place less stringently. For example, in France and the United Kingdom, compliance with the limit value or a future limit value is one of the factors considered in the permit process, but this compliance can be made subsidiary to other societal interests. Although the limit values are strictly enforced in Germany, the consequences that result from threatened non-compliance have, until now, been less far-reaching than those in the Netherlands (Koelemeijer *et al.*, 2005).

In the Netherlands, the limit values apply everywhere in the country, regardless of whether there is actually any exposure to people. In other EU countries, the limit values theoretically apply to the entire country, but at least in Germany and Austria, the law is interpreted in such a way that the limit values only apply to locations where people could be affected. It is clear that the European legislation leaves space for various interpretations at the national level (Koelemeijer *et al.*, 2005).



### 3. How high is the emission?

- Between 1990 and 2003, the primary particulate matter emission in the Netherlands declined by 50%. The emissions of precursors of secondary particulate matter – ammonia, nitrogen oxides and sulphur dioxide – also declined sharply during this period.
- In most other EU 25 countries, the emission of primary particulate matter also declined. This decline was frequently the result of comparable European and national policies, and the measures that emerged from these policies.
- Between 2000 and 2020, it is expected that the particulate matter emission in the Netherlands will decline slightly or remain constant. During this period, emission from traffic will decline by 25%. Emissions will also decline in all other countries of the European Union.
- The emissions in other EU countries will decline more quickly in the future than those in the Netherlands. This is because the Netherlands has already implemented a relatively large number of control technologies. In addition, there has been a virtually complete transition to natural gas.

#### *Overview of the chapter*

This chapter addresses the emissions of primary particulate matter in the Netherlands and a number of other European countries. It also discusses the expected developments in emissions, the uncertainties in these expectations and the effects of measures to control emissions.

#### **Emissions in the Netherlands**

Every year, the Dutch Emission Inventory records the emissions of primary particulate matter in the Netherlands. Primary particulate matter is particulate matter that is emitted directly into the atmosphere. The relevant authority, usually the province, monitors the emissions that are reported by large companies. The emissions from other sectors, including traffic, consumers, agriculture, trade, services and government, is calculated by sector committees. All these committees operate within the Emission Inventory.

The level of uncertainty in the monitoring of total particulate matter emissions is not well known (MNP, 2005). A recent study by the Netherlands Organization for Applied Scientific Research (TNO) showed that the uncertainty in monitoring emissions from the known sources is at least 20% (TNO, 2004). Until now, the Emission Inventory has not estimated  $PM_{2.5}$  emissions; its figures for particulate matter are expressed only as  $PM_{10}$ . Particulate matter that is directly emitted from combustion processes, such as transport, industry and consumers, is composed of particles that are also smaller than  $PM_{2.5}$ . Particulate matter that is emitted from mechanical processes, such as road wear and emissions from animal husbandry, primarily involves particles that are larger than  $PM_{2.5}$ .

## Emissions in other European countries

As part of the Convention on Long-range Transboundary Air Pollution, data about the emission of primary particulate matter in other European countries must be reported annually to the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP). Until now, few countries have met this obligation. In 2004, the Netherlands was one of 16 out of 50 countries that met its EMEP obligation to report on 2002 emissions (EMEP, 2005a). For the other countries, EMEP estimates the magnitude of their annual emissions (Vestreng, 2004). The uncertainty in the particulate matter emission data from other European countries is also not well understood (EEA, 2003). For the time being it is impossible to quantify this uncertainty. Prognoses for particulate matter emission are made for Europe using the RAINS model. This is the air pollution model that the European Commission uses to support new air pollution policy.

## Emission trends in the Netherlands

Between 1990 and 2003, the emission of primary particulate matter in the Netherlands declined by nearly 50% (*Table 3.1*). The largest declines occurred with companies and road traffic (MNP, 2005). The emissions of precursors of secondary particulate matter – ammonia, nitrogen oxides and sulphur dioxide – also declined sharply during this period (by 48%, 32% and 66%, respectively). The declining emissions of primary particulate matter in the Netherlands from companies (industry, refineries and the energy sector) is primarily due to legislation such as the Order Governing Combustion Plant Emission Requirements and the Netherlands Emission Regulations. This has led to measures such as process modifications and more widespread use of filters. The decline from traffic is due to European legislation on exhaust emissions.

*Table 3.1 Emission of primary particulate matter in the Netherlands, 1990-2003<sup>a</sup>.*

Emission per sector	1990	1995	2000	2002	2003	2010
<i>Millions of kg</i>						
Industry, energy sector and refineries	38	23	13	13	12	12
Traffic	23	20	17	16	16	13
a. Of which road traffic	18	14	12	11	11	9
of which diesel vehicles <sup>b</sup>	14	10	8	7	6	6
b. Of which wear <sup>c</sup>	3	3	3	3	3	4
Consumers	4	4	4	4	4	9 <sup>d</sup>
Trade, services, government and construction	4	3	4	4	3	
Agriculture	9	10	10	9	8	10
<b>Total PM<sub>10</sub></b>	<b>78</b>	<b>59</b>	<b>49</b>	<b>45</b>	<b>42</b>	<b>44</b>

a) The emissions from shipping are not included in this table. In 2000 these amounted to 2 million kg for emissions in ports and 8 million kg for emissions on the continental portion of the Netherlands. (Emission Inventory, 2005).

b) 30% originates from automobiles, 70% from trucks (including delivery vans and busses).

c) Wear from tyres, road surfaces and brakes.

d) Total consumers, trade, services, government and construction.

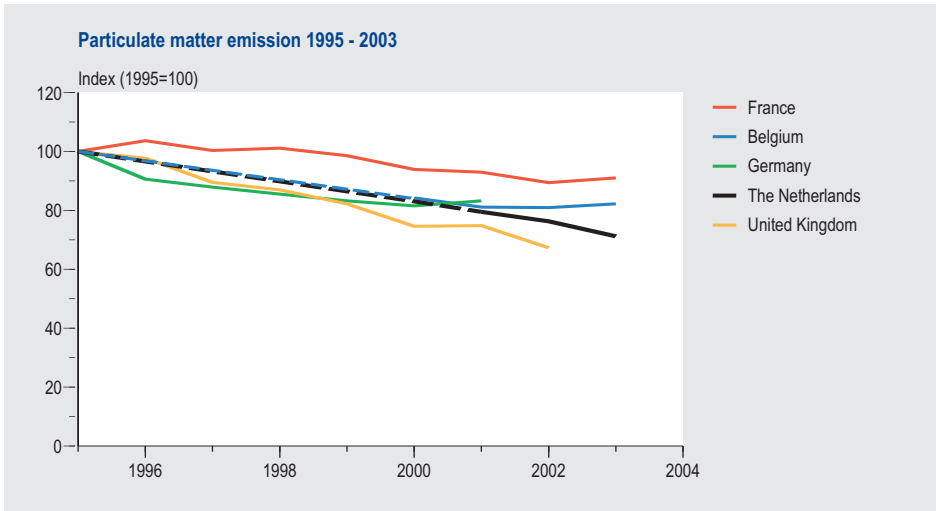


Figure 3.1 Emission of primary particulate matter in the Netherlands and a number of neighbouring countries, 1995-2003. For the Netherlands during this period, data are only available for 1990 and 2000. Source: Umweltdaten Duitsland, 2005; MIRA, 2005; Citepa, 2005; NAEI, 2005.

## Emission trends in other European countries

In most of the other EU 25 countries the emission of primary particulate matter also declined. This decline was primarily due to comparable European and national policies and the measures that emerged from these policies. In Germany and in many of the recently-admitted EU countries, the decline also resulted from the closure of brown coal power plants and the shift to other fuels such as natural gas. The closure of unprofitable factories also contributed to the decline in emissions. However, it is unclear exactly how great this decline has been in recent years. This is because, as indicated above, only a few EU 25 countries submit reports on emissions. The trend in emissions of primary particulate matter in the Netherlands and its four neighbouring countries during the period 1995-2003 is shown as an example in *Figure 3.1*.

As in the Netherlands, the emissions of particulate matter precursors also declined in the EU 25. Between 1990 and 2002, the decline for ammonia was 16%, for nitrogen oxides 31% and for sulphur dioxide 66% (EEA ETC/ACC, 2004).

## Future emissions

Depending on the scenario, particulate matter emissions in the Netherlands are expected to decline slightly (-15%) or remain constant between 2000 and 2020 (ECN/MNP, 2005). During this same period, emissions caused by traffic will decline by 25%. There will be little change in the emissions caused by other target sources (*Table 3.1*). According to calculations with the RAINS model (RAINSb, 2005), in the rest of Europe (EU 25) the future anthropogenic emissions of particulate matter, in the form

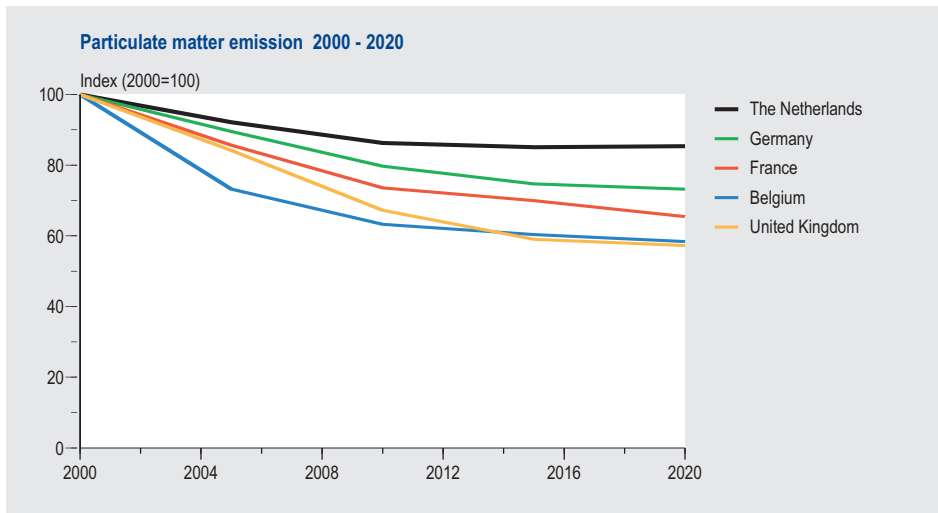


Figure 3.2 Emission of primary particulate matter between 2000-2020 in Germany, Belgium, France, the United Kingdom and the Netherlands, according to the RAINS model. The expectation is that the emissions in other countries will decline more rapidly than in the Netherlands. This is because a relatively large number of control technologies have already been applied in the Netherlands. The fact that there has been almost a complete transition to natural gas in the Netherlands also plays a role.

of both  $PM_{10}$  and  $PM_{2.5}$ , will also decline. However, the emissions in other countries will decline more rapidly than in the Netherlands (Figure 3.2). This is because, as stated previously, other countries will begin to catch up with the Netherlands, which has already implemented a relatively large number of control technologies and has made a virtually complete transition to natural gas.

For the RAINS calculations, data and scenario assumptions are used for each country concerning economic development, the number of residents, the energy use, the total distance travelled by vehicles, the number of animals in agriculture, the industrial production, emission factors and the application of emission control measures. This scenario deviates somewhat from the above scenario in the Netherlands. The RAINS input data, as part of the Clean Air for Europe (CAFE) programme, was checked and improved by the relevant countries in 2004. The Netherlands data has been checked for both  $PM_{10}$  and  $PM_{2.5}$  (Jimmink, 2004). For the Netherlands, RAINS calculates a higher particulate matter emission for the year 2000 – 15% to 20% higher – than shown in the data from the Dutch Emission Inventory. This will also be the case in the future. This deviation is due to scenario differences regarding aspects such as the numbers of livestock and fuel consumption by consumers. The comparison of the currently available data from 2000 for particulate matter in RAINS indicates that a number of countries have failed to sufficiently check their RAINS data for 2004 as well. For example, the emission factors for many emission sources are identical in all countries. Moreover, during the autumn of 2005, the data will be once again checked by the countries as part of the revision of the National Emissions Ceilings Directive (NEC).



## 4. How much particulate matter do we measure?

- The measured annual average concentration of particulate matter in 2003 was about  $34 \mu\text{g}/\text{m}^3$ . That is 25% lower than 10 years ago.
- In the Netherlands, the limit value for the annual average concentration and that for the 24-hour average are both being exceeded. Measurements show that the limit value for the 24-hour average is exceeded more often than that of the annual average concentration. The limit value for the 24-hour average therefore appears to be more stringent than the limit value for the annual average concentration.
- Particulate matter concentrations are measured in the Netherlands according to a methodology prescribed in European legislation. Measurements of  $\text{PM}_{10}$  are conducted in the Netherlands at 39 locations; 22 of these locations are in urban surroundings.
- Components of particulate matter are: inorganic secondary components, components that contain carbon, sea salt, oxides of metals and silicon and water. Sea salt and soil dust are important components of particulate matter; on an annual average basis, they amount to 20% to 30% of total particulate matter.
- Meteorological influences can lead to fluctuations in the annual average particulate matter concentration of around  $5 \mu\text{g}/\text{m}^3$ .
- Subtracting the contribution of sea-salt aerosol from the total particulate matter concentration has little effect on how often the limit value for the 24-hour concentration is exceeded. On average for the Netherlands, it is estimated that subtracting sea-salt aerosol results in six fewer days when the limit value for the 24-hour average is exceeded.

### *Overview of the chapter*

Chapter 4 addresses the measured concentrations of particulate matter in the Netherlands, the fact that the two European limit values are exceeded and the relationship between these limit values. In addition, this chapter provides information about the infrastructure used for measurements, about the measurements themselves and about the measured components of particulate matter.

### **Concentrations in the Netherlands**

The air quality regarding particulate matter in the Netherlands has improved during the past decade. In 2003, the measured annual average concentration of particulate matter was  $34 \mu\text{g}/\text{m}^3$ . The annual average concentrations have declined by 25% in ten years. During the same period, the number of days with a 24-hour average concentration above  $50 \mu\text{g}/\text{m}^3$  declined by 50%. Nevertheless, both limit values are still exceeded in the Netherlands. It appears that the limit value for the 24-hour average is exceeded more often than the limit value for the annual average concentration (*Figure 4.1*, *Figure 4.2*). Future developments will be discussed in greater detail in Chapter 5, *How much particulate matter do we calculate?*

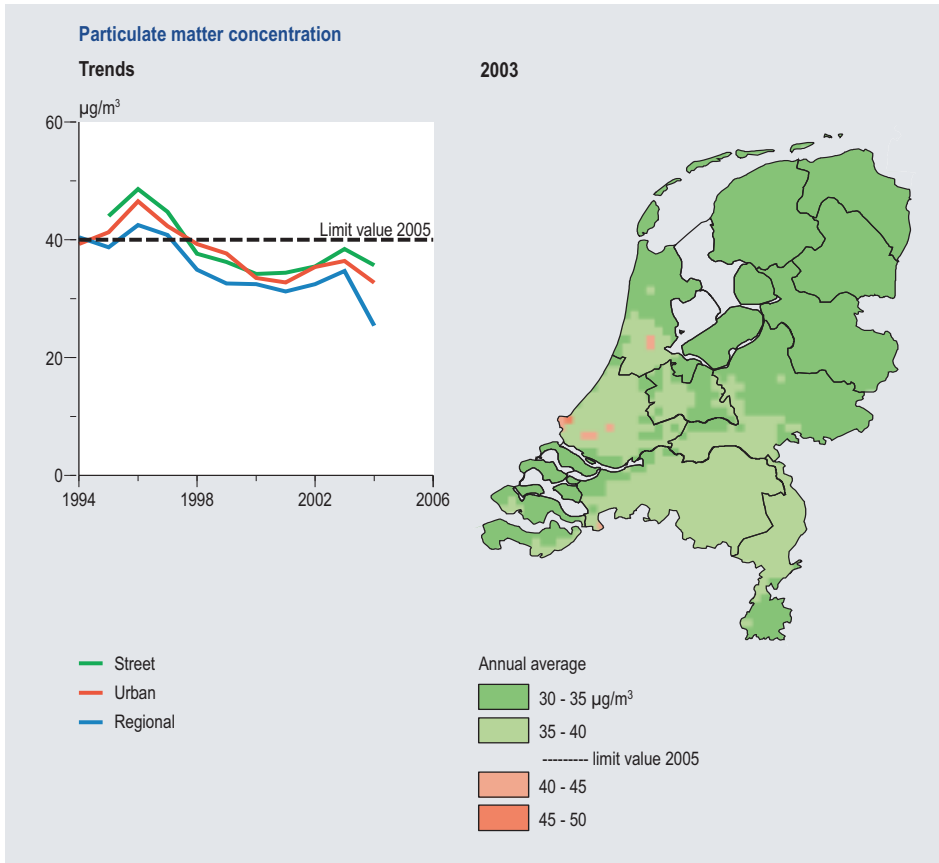


Figure 4.1 Measured annual average particulate matter concentrations in the Netherlands in 2003. The trend lines (left) indicate the average of the stations in the corresponding group. The map for the annual average concentrations was obtained from measurement results from the Dutch National Air Quality Monitoring Network combined with model calculations; for an explanation, see Chapter 5, 'How much particulate matter do we calculate?' Source MNC, 2005.

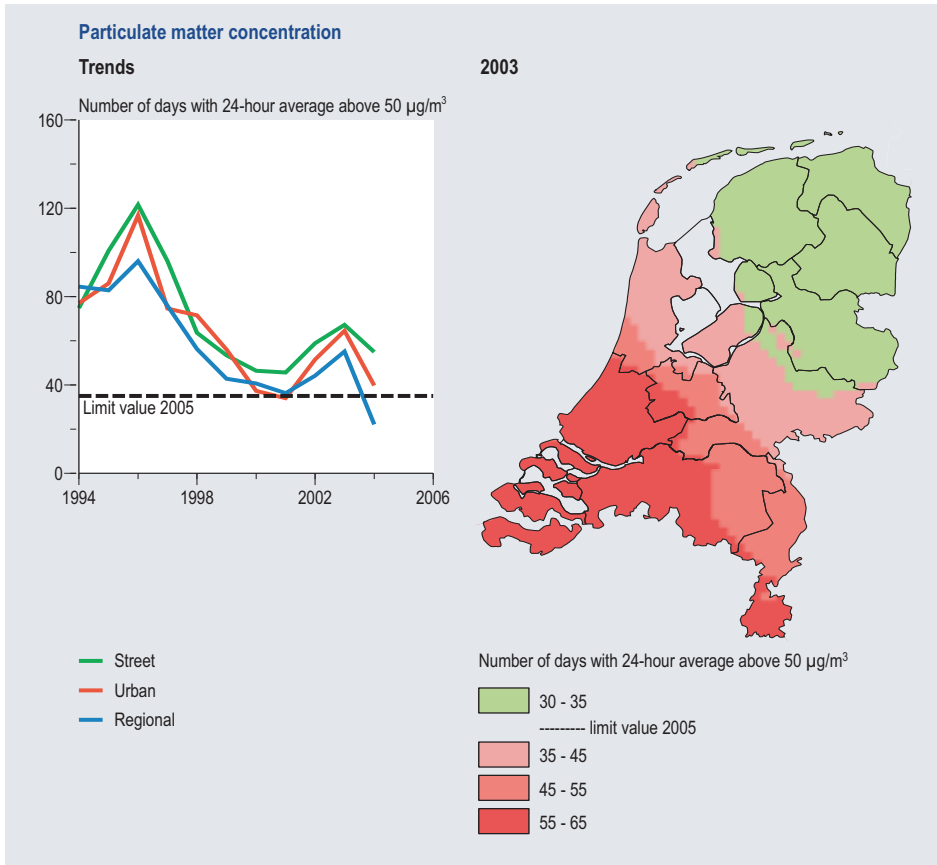


Figure 4.2 The number of days with a 24-hour average above  $50 \text{ mg}/\text{m}^3$  in the Netherlands in 2003. The trend lines (left) give the average of the stations in the corresponding group. The map for the number of days the limit value was exceeded was arrived at through interpolation of the measurement results from the regional measurement stations in the Dutch National Air Quality Monitoring Network. The compliance problems concerning the limit value for the 24-hour average concentration occur over a large part of the Netherlands (right). Source: MNC, 2005.

The measurement results for particulate matter also show the relationship between the two European limit values: the limit value for the annual average concentration,  $40 \mu\text{g}/\text{m}^3$ , and the limit value for the 24-hour average; the latter is a maximum of 35 days per year with a 24-hour average concentration above  $50 \mu\text{g}/\text{m}^3$  (Figure 4.3). This relationship shows that the limit value for the 24-hour average corresponds with an annual average particulate matter concentration of approximately  $31 \mu\text{g}/\text{m}^3$ . The limit value for the 24-hour average is therefore significantly more stringent than the limit value for the annual average concentration.

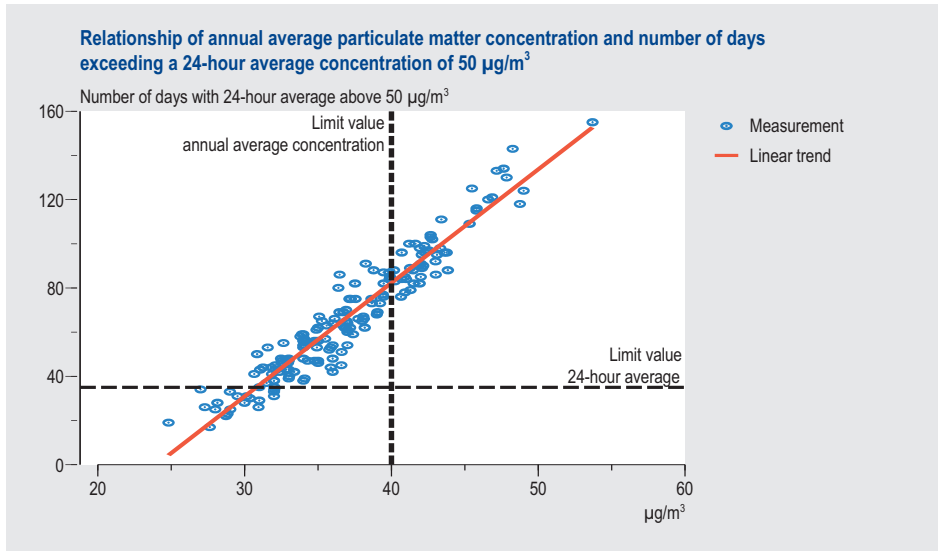


Figure 4.3 Relationship between the annual average particulate matter concentration and the number of days with a 24-hour average above  $50 \mu\text{g}/\text{m}^3$ . The vertical line indicates the limit value for the annual average concentration. The horizontal line indicates the maximum number of days permitted with a 24-hour average concentration above  $50 \mu\text{g}/\text{m}^3$ . From this relationship it follows that every additional microgram of particulate matter results in five more days that exceed the limit value for the 24-hour average concentration. According to this relationship, at an annual average concentration of  $31 \mu\text{g}/\text{m}^3$ , neither limit value will be exceeded. In that case there are precisely 35 days with a 24-hour average concentration of  $50 \mu\text{g}/\text{m}^3$ .

## Trends in the concentration

Meteorological year-to-year fluctuations have a clear influence on the annual average particulate matter concentration in the Netherlands (Figure 4.4). However, it is possible to correct for these fluctuations (Visser and Noordijk, 2002). After such a meteorological correction is made, it appears that between 1992 and 2003 a downward trend of  $1 \mu\text{g}/\text{m}^3$  per year occurred on average. In addition, the number of days that exceeded the limit value for the 24-hour average also declined on average during the same period.

Meteorological influences can lead to fluctuations in the annual average particulate matter concentration of around  $5 \mu\text{g}/\text{m}^3$  (Figure 4.4). This means that if the Netherlands intends to comply with the limit value for the annual average concentration of  $40 \mu\text{g}/\text{m}^3$  for each individual year, the concentration must lie around  $35 \mu\text{g}/\text{m}^3$  during a meteorologically normal year. If the Netherlands also intends to comply with the limit value for the 24-hour average, then the annual average concentration cannot be more than  $26 \mu\text{g}/\text{m}^3$  (Figure 4.3).

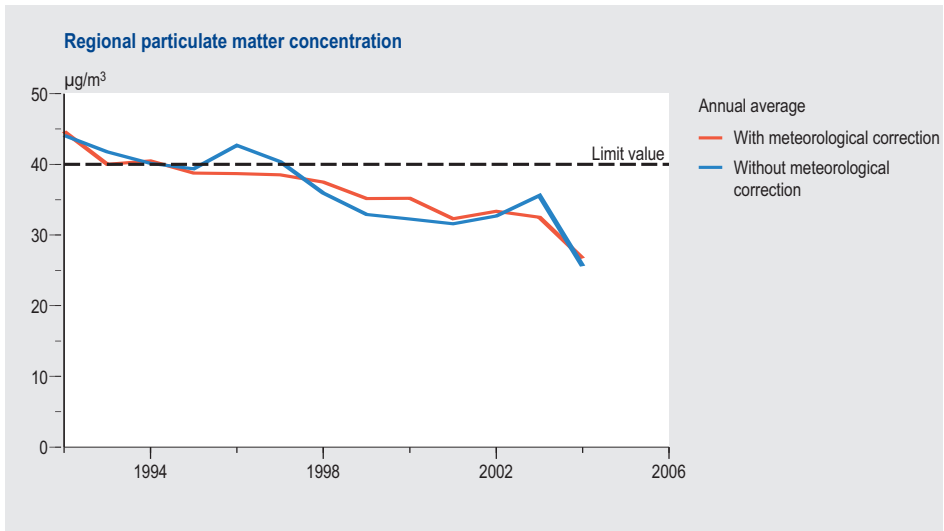


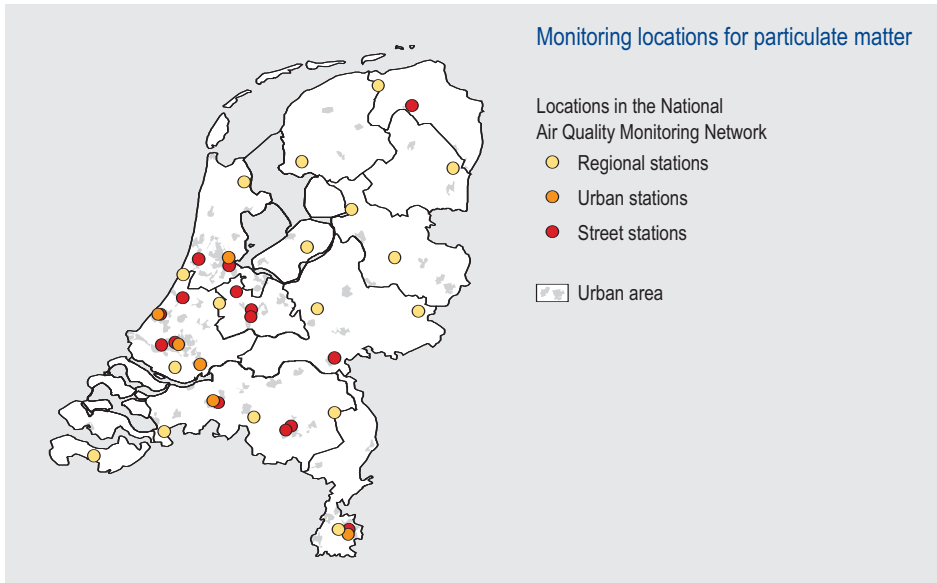
Figure 4.4 Measured trend in the annual average particulate matter concentration at the regional stations of the Dutch National Air Quality Monitoring Network. The figure shows the trend both with and without correction for coincidental fluctuations in meteorology. During a year with non-beneficial meteorological conditions, the annual average particulate matter concentration can be up to  $5 \mu\text{g}/\text{m}^3$  higher than during a year with normal meteorological conditions.

## Monitoring infrastructure

The monitoring of particulate matter concentrations takes place in the Netherlands according to a methodology prescribed in European legislation. This monitoring has taken place in the Netherlands since 1992 (Van Elzakker, 2001). In terms of monitoring, the Netherlands is therefore one of the leaders in the European Union, together with Finland and the United Kingdom (Buijsman *et al.*, 2004). Initially, there were 19 monitoring stations in the monitoring network for particulate matter in the Netherlands.

The Netherlands has chosen the numbers of monitoring stations in such a way that it is possible on the basis of the measurement results alone – therefore without the use of models – to provide a representative picture of the air quality for particulate matter in the Netherlands. However, to ensure that the monitoring network meets the requirements of European legislation, the network configuration and the number of monitoring stations had to be revised. In mid-2005, this revision had not yet been fully completed. On 1 July 2005, there were 17 regional stations, 6 urban stations and 16 street stations in the monitoring network for particulate matter. Two more street stations are planned. This will bring the total number of monitoring stations for particulate matter up to the intended number of 41 (Figure 4.5).

In the Netherlands, there are also a number of regional and local authorities which monitor particulate matter. The provinces of Limburg and North Holland have moni-



*Figure 4.5 Monitoring locations for particulate matter ( $PM_{10}$ ) in the Dutch National Air Quality Monitoring Network, mid-2005. Regional and local monitoring networks for particulate matter are not shown. Regional station: station that is located outside the built-up area and is not affected by local sources. The spatial representativity is assumed to be such that by taking the results of all these stations together, a picture of the entire country can be made. Urban station: a station in an urban area that is located in such a way that fewer than 2,750 vehicles pass within 35 metres of the station each day (Eerens et al., 1993). Street station: a station in an urban area that is located in such a way that at least 10,000 vehicles pass within a radius of 35 metres around the station each day (Eerens et al., 1993). Two street stations are not yet operational and are not shown on the map. This concerns street stations in Amsterdam and The Hague. Source: Laboratory for Environmental Monitoring /RIVM.*

toring networks with two and six monitoring stations, respectively. The monitoring network of the DCMR Environmental Protection Agency has three monitoring stations, and the monitoring network of the GG & GD in Amsterdam comprises five monitoring stations. Particulate matter measurements are also conducted near the Corus steel mill in Wijk aan Zee.

## Monitoring apparatus

The particulate matter measurements are conducted in the Netherlands with automatic monitoring apparatus that works according to the principle of the attenuation of beta radiation. A widely-used term for a device of this type is a beta attenuation analyser. The operation of this type of equipment is not based on an absolute measurement method. Moreover, the method has measurement artefacts that must be corrected (see text box *The measurement of particulate matter and correction factors*).

### The measurement of particulate matter and correction factors

A reference method was prescribed in the first EU daughter directive for measuring particulate matter where particles in the air are trapped in a filter; the mass of the particulate matter is then determined by weighing. However, this method is very time consuming. The directive also permits the use of an alternative method if it can be shown that the results are sufficiently comparable with the reference method. The most widely-used alternative methods are the beta attenuation method and the TEOM method (see below). The result of the measurements made with these automatic instruments are affected by the temperature of the air that is drawn into the instruments and the temperature of the filter. This temperature setting is a compromise between preventing condensation on the one hand and limiting the evaporation of volatile aerosols on the other. As a result, both the beta attenuation method and the TEOM method have a systematic deviation with respect to the reference method; these deviations are often corrected. A correction factor of 1.33 is used on the measurement results of the beta attenuation monitors in the Dutch National Air Quality Monitoring Network.

The beta attenuation method uses beta radiation (beta particles) for the measurement. This radiation is attenuated when it is transmitted through solid matter. The beta attenuation analyser uses a filter band through which the beta particles are transmitted. The beta particles are measured before and after the filter is loaded with particulate matter. The degree of attenuation correlates with the quantity of particulate matter on the filter and consequently with the particulate matter concentration in the air. The entire cycle of zero-point calibration, sampling and measurement is automated.

TEOM stands for Tapered Element Oscillating Microbalance. This method uses a tapered glass element on which a filter is located. This element oscillates with a characteristic frequency. Loading the filter with particulate matter leads to a change in the oscillation frequency. The degree of change correlates with the quantity of particulate matter on the filter and consequently with the particulate matter concentration in the air. The entire cycle of zero-point calibration, sampling and measurement is also automated with this method.

The so-called semi-volatile components are a problem with both the reference method and the automatic monitoring methods for particulate matter. The magnitude of the problem depends

partly on the temperature settings in the relevant apparatus. This concerns both organic and inorganic components. In this context, ammonium nitrate ( $\text{NH}_4\text{NO}_3$ ) is an especially important component. Losses of this component of particulate matter that have been collected on a filter occur because ammonium nitrate is in balance with ammonia ( $\text{NH}_3$ ) and nitric acid ( $\text{HNO}_3$ ). The balance can change during the sampling procedure. During a 24-hour period, this can lead to unpredictable losses of ammonium nitrate.

Moreover, to prevent condensation of moisture, the suction tube is heated until just above the location of the filter. However, this leads to losses of semi-volatile components in the particulate matter. To compensate for these losses, a correction factor is applied. The value of the correction factor must be ascertained by means of comparative research, where the results of the automatic method are compared with the results according to the reference method. Many measurements of particulate matter concentrations take place with the automatic apparatus referred to above. Both the use of this apparatus and the application of the correction factors are explicitly allowed under European legislation. However, this must take place under the condition that equivalence with the reference method for measuring particulate matter has been shown.

At the beginning of the 1990s, a correction factor of 1.33 was ascertained in the Netherlands based on research that took place at that time. Later on, Dutch research showed that the correction factor probably differs in time and space (Van Putten *et al.*, 2002). At the same time it was ascertained that the correction factor for urban stations was possibly too low and that for regional stations was possibly too high. Another study on this topic is currently taking place in the Netherlands. If they do not have any results from their own research, a number of countries follow the recommendation of the European Commission and use a correction factor of 1.3. Only Belgium uses a higher correction factor than the Netherlands. Most countries use a lower correction factor or sometimes no correction factor at all. This is often based on research conducted by the countries themselves, although it is unclear for a number of countries how they arrived at a value for the correction factor (Buijsman and De Leeuw, 2004). Recent research has again provided indications about the location dependency and season dependency of the correction factor (Heldstab and Stampfli, 2001).

The calculated uncertainty in the 24-hour average measured particulate matter concentration is 15%. The calculated uncertainty in the annual average concentration is 9% (Blank, 2001). In the first daughter directive, the European Union required a maximum uncertainty of less than 25% for both measurements (EU, 2001). The measurements in the Netherlands are therefore well below this uncertainty limit. However, it should also be noted that the indicated percentages are probably minimum values, because not all factors that can influence the uncertainty can be quantified.

Before 1992, particulate air pollution was monitored in the Netherlands only in the form of black smoke (Van Elzakker, 2001). However, black smoke is only a portion of particulate matter (and  $PM_{2.5}$ ). Therefore, statements about a possible trend in particulate matter concentrations can only be made on the basis of data from 1992 onwards.

## Components of particulate matter

The measurement of particulate matter with the beta attenuation method is given in terms of mass per unit volume. It does not provide any information about the chemical composition of the particulate matter. Research has shown that the most important components of particulate matter are inorganic secondary components, carbon-containing components, sea salt, oxides of metals and silicon and water (Visser *et al.*, 2001; see the text box *Chemical composition of particulate matter in the Netherlands* for a more extensive explanation).

Sea salt and soil dust are important components of particulate matter: on a yearly average, they amount to 20% to 30% of total particulate matter. Between 25% and 50% of the sea salt aerosol is composed of particles in the  $PM_{2.5}$  fraction (Visser *et al.*, 2001). Sea salt and the natural component of soil dust cannot be influenced by policy measures. Moreover, it is very probable that sea salt does not have any health effects. In current European legislation for particulate matter, however, the *total* concentration, therefore including fractions of natural origin, is regulated. As a result, a discussion is now taking place in the Clean Air for Europe (CAFE) programme about the possibility of making a statutory exception for components in particulate matter of natural origin which are also viewed as non-hazardous.

An important question is: what is the relationship between the concentrations of sea salt and soil dust on the one hand and non-compliance with the limit values on the other? To answer this question, an initial estimate of the long-term average contribution of sea-salt aerosol to the particulate matter concentration in the Netherlands was made. This contribution was estimated at 4 - 5  $\mu\text{g}/\text{m}^3$ . Measurements have shown that the annual average sea-salt aerosol concentration on the coast is between 5 and 8  $\mu\text{g}/\text{m}^3$ ; in the southern province of Limburg and on the eastern border of the Netherlands, this value is 3  $\mu\text{g}/\text{m}^3$  (Figure 4.6; Visser *et al.*, 2001; Denier van der Gon *et al.*, 2003). Measurements of sea salt aerosol in the German federal state of Nordrhein-Westfalen indicate an average concentration of 1 to 2  $\mu\text{g}/\text{m}^3$ . These figures support the assumed distribution across the Netherlands.



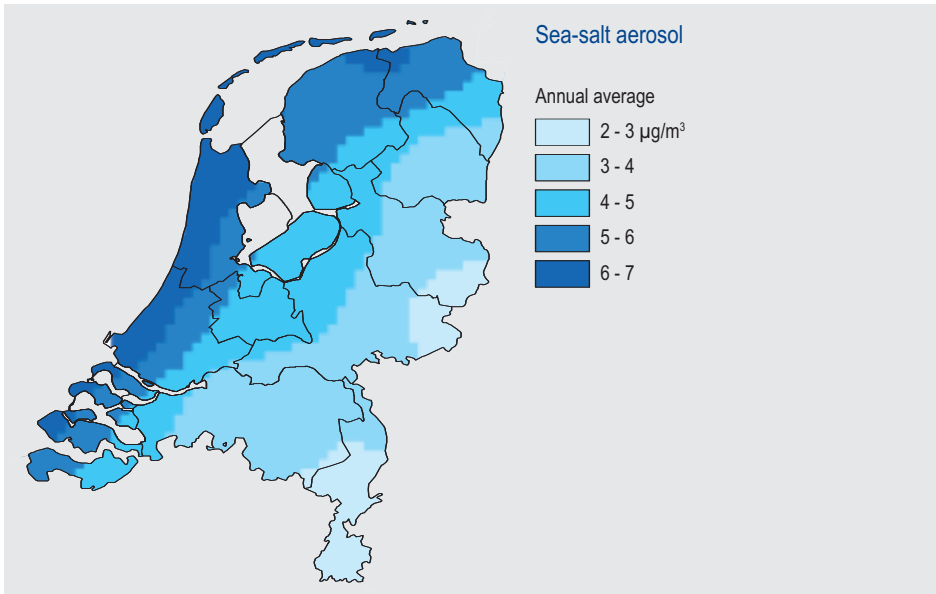


Figure 4.6 Annual average contribution of sea salt aerosol to the particulate matter concentration in the Netherlands. The estimate is based on interpolation of monitoring results and has been combined with assumptions about the distribution of sea salt along the Dutch coast (Eerens *et al.*, 1998; Eerens, 1998).

Tentative estimates indicate that subtracting the contribution of sea-salt aerosol would bring the particulate matter concentration below the limit value for the annual average concentration of  $40 \mu\text{g}/\text{m}^3$  (under average meteorological conditions). It is expected that this limit value would then only be exceeded locally, due to local traffic contributions.

Non-compliance with the limit value for the 24-hour average is the most acute problem concerning the air quality regulations for particulate matter. For the year 2020, it has been calculated that there will still be between 50 and 70 instances where the limit value is exceeded at the highway ‘hotspots’ and in the inner cities that were studied (Beck *et al.*, 2005b). However, it is expected that subtracting the contribution of the sea-salt aerosol will have little effect on bringing the Netherlands into compliance with the limit value for the 24-hour average. This is because high particulate matter concentrations generally occur due to air movement from continental Europe. Under such conditions, the share of sea-salt aerosol in the particulate matter concentration is quite small (Matthijssen, 2005; Denier van der Gon and Schaap, 2005). The relationship between exceeding the limit value for the 24-hour average particulate matter concentration and the concentration of sea-salt aerosol has been investigated based on several measurements. It is estimated that subtracting the sea-salt aerosol will lead, on average for the Netherlands, to six fewer days when the value for the 24-hour average is exceeded. This estimate has a 50% uncertainty.

### Chemical composition of particulate matter in the Netherlands

Recent measurements of the chemical composition of particulate matter in the Netherlands have led to a good understanding of the average composition of particulate matter (Visser *et al.*, 2001). The components are the following:

**Inorganic secondary components.** This primarily concerns sulphate (SO<sub>4</sub>), nitrate (NO<sub>3</sub>) and ammonium (NH<sub>4</sub>) aerosol. These particles are formed in the atmosphere from the gases sulphur dioxide (SO<sub>2</sub>), nitrogen oxides (NO<sub>x</sub>) and ammonium (NH<sub>3</sub>). The contributions in terms of mass of these components to the annual average concentration is around 10 µg/m<sup>3</sup>. Approximately 90% of these components are found in the PM<sub>2.5</sub> fraction. Virtually all these secondary components are of anthropogenic origin.

**Carbon-containing components.** This primarily concerns elementary carbon and organic compounds. The majority of these compounds are emitted directly into the atmosphere. The contribution in terms of mass of these components is 5 µg/m<sup>3</sup>, of which 1 µg/m<sup>3</sup> is elementary carbon and 4 µg/m<sup>3</sup> comprises organic compounds. Soot is composed of a mixture of elementary carbon and organically bound carbon. A small portion of the carbon-containing components is formed in the air by a chemical reaction; this is the secondary organic aerosol. Of the carbon-containing components, 90% are found in the PM<sub>2.5</sub> fraction. This group also contains a very small (in terms of mass) quantity of polycyclic aromatic hydrocar-

bons (PAHs). The elementary carbon and the PAHs are entirely of anthropogenic origin, while the other organic compounds are partly of natural origin and partly of anthropogenic origin.

**Sea salt.** Sea salt is composed primarily of sodium chloride (table salt) with a smaller contribution from magnesium, calcium and potassium compounds. Sea-salt aerosol is of natural origin and is formed in the air when the wind blows over the sea's surface. On average across the Netherlands, the contribution in terms of mass of sea salt is 4-5 µg/m<sup>3</sup>. Between 25% and 50% of the sea salt is found in the PM<sub>2.5</sub> fraction.

**Oxides of metals and silicon.** This primarily concerns oxides of silicon, aluminium, calcium, iron and potassium. This can be largely attributed to windblown soil dust. This soil dust comes into the air primarily as the result of human activities. The total soil dust concentration is on the order of 4 µg/m<sup>3</sup> on average across the Netherlands. Soil dust is largely comprised (70% to 90%) of particles larger than PM<sub>2.5</sub>.

**Water.** Components of particulate matter, especially inorganic secondary components, can contain chemically-bound water. The contribution of chemically-bound water is estimated at 10% to 15% of the total particulate matter concentration. The share of water in the PM<sub>2.5</sub> fraction is larger because water is bound especially to inorganic secondary components.

## 5. How much particulate matter do we calculate?

- The annual average concentration of particulate matter in 2003 was  $34 \mu\text{g}/\text{m}^3$ . Of this concentration, 45% was calculated based on anthropogenic emissions (on average for the Netherlands). All particulate matter models have this phenomenon in common. Two-thirds of the anthropogenic portion comes from outside the Netherlands and one third originates from inside the country. The other portion originates primarily from natural sources, but also sources of which the magnitude is unknown or has been possibly incorrectly estimated.
- The primary sources for the concentration of particulate matter in the Netherlands are traffic and agriculture in the Netherlands itself and traffic and industry in other countries.
- At least 15% of the total concentration originates from anthropogenic sources in the Netherlands. Locally, in urban areas, this level is higher (30%-45%), especially due to traffic. It is virtually impossible to comply with the limit values for particulate matter everywhere in the Netherlands by means of national policy alone.
- Due to its high emission density, however, the Netherlands is a net exporter of particulate matter. The export of particulate matter from the Netherlands is three times as large as the import.
- During the past ten years, concentrations have fallen by 25%. This trend will continue in the future, although to a lesser degree. It is especially emission reductions abroad that contributed to this decline. Nevertheless, in the near future the limit value for the 24-hour average will continue to be exceeded on a large scale.
- Calculations show that the main 'hotspots' in Europe, besides the Netherlands, are Belgium, the Ruhr region in Germany and North Italy.

### *Overview of the chapter*

This chapter addresses the use of models in the analysis of air quality for particulate matter. A description of the methodology of the model calculations is provided. Causes and differences between models and differences with the results of measurements are discussed.

### **The role of model calculations**

Model calculations are used to evaluate and explore environmental policy, and they are essential to the interpretation of measurement data. In addition, the Netherlands has chosen to use models to ascertain air quality and report on this air quality to the European Commission. Models are also used to review construction plans in terms of potential compliance with air quality limit values as part of the permit process. In preparing policy for Europe, models are used that calculate the air quality for the entire European land area (RAINS, 2005; EMEP, 2005b). The RAINS model makes integrated air quality evaluations across the entire chain, from source to effect and the reverse. Data calculated by the EMEP dispersion model are the basis input for the RAINS model and therefore play an important role in the policy formation process in

the European Union. The instrumentation used in the Netherlands provides a much higher resolution than the EMEP model, but the results are limited to the air quality in the country itself. This situation plays a role, for example, in determining the Dutch standpoint in Brussels.

Particulate matter can remain in the atmosphere for days; as a result it can be transported across thousands of kilometres. Sources far from the Netherlands therefore contribute to the concentrations in the Netherlands. Local sources also contribute to the concentrations in the country. The calculation methodology is described separately in this chapter (see text box *Methodology for calculating particulate matter concentrations*).

## Background concentration in the Netherlands

Figure 5.1 shows the concentrations of particulate matter as they are calculated based on emissions for the years 2000 and 2010. It is obvious that the concentrations are

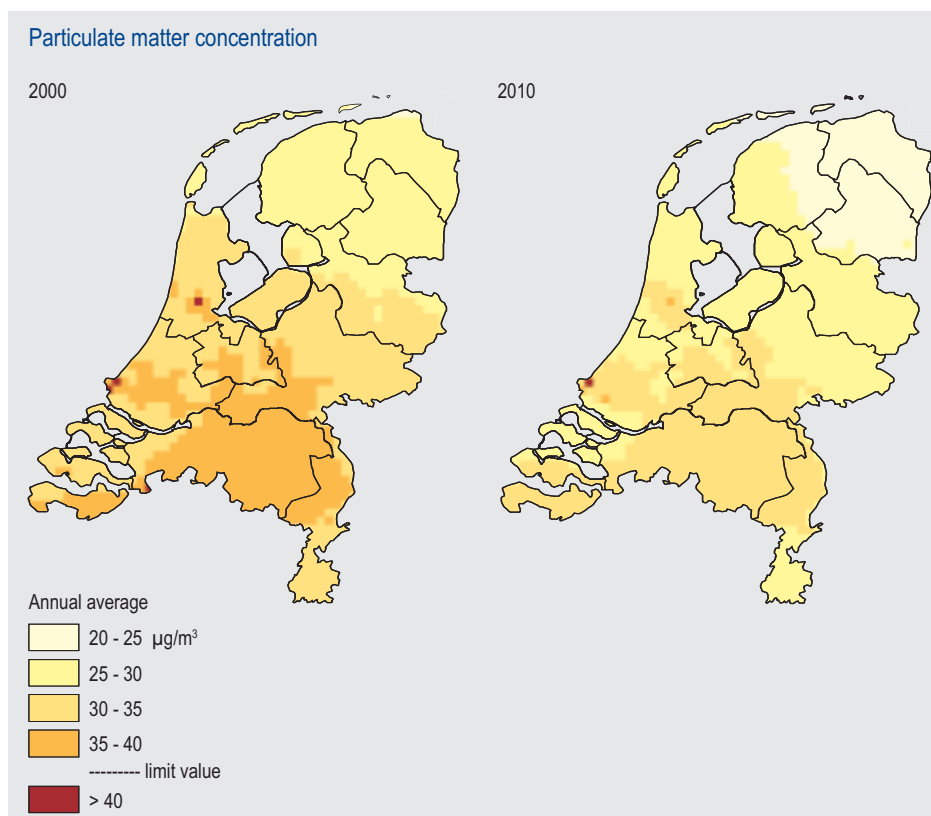


Figure 5.1 Calculated annual average concentration of particulate matter in 2000 (left) and 2010 (right), based on ascertained and/or expected emissions and many-year average meteorological conditions (1990-1999). The calculations for 2000 are 'calibrated' to measurements. The calculations for 2010 have been made based on the CAFE baseline scenario for countries outside the Netherlands and the Global Economy Reference Framework for the Netherlands (Folkert et al., 2005).

higher in the urban agglomerations of the western part of the country and in the southern provinces than in the northern part of the Netherlands. This is due to the higher emissions from traffic and business activities in these regions. The proximity of sources in other countries, especially in Belgium and the Ruhr district of Germany, also plays a role. According to expectations, in 2010 it is possible that the limit value for the annual average concentration will still be exceeded locally in some parts of the Netherlands. The limit value for the 24-hour average will probably still be exceeded at that time on a large scale. As shown in *Figure 4.3*, the maximum number of days when the 24-hour average particulate concentration is above the limit value will be exceeded at locations where the annual average concentration is above  $31 \mu\text{g}/\text{m}^3$  (*Figure 4.3*).

Since the 1990s, the background concentration of particulate matter has dropped by  $1 \mu\text{g}/\text{m}^3$  per year. Calculations show that three-fourths of this decline is due to emission reductions in other countries. Forty percent of the decline is due to the reductions in emissions of primary particulate matter and 60% is due to the decline in the emissions of precursors of secondary particulate matter. After 2005, the concentration will continue to decline under the influence of the established European and national policy. However, the decline between 2000 and 2010 will flatten out and is estimated at about  $0.5 \mu\text{g}/\text{m}^3$  per year on average across the Netherlands. During this same period, an estimated 85% of the decline in concentration in the Netherlands will be the result of emission-reducing measures abroad. This is logical, because the background concentration in the Netherlands is dominated by contributions from abroad. It is especially source measures in industry and traffic that will contribute to this decline.

Of the average particulate matter concentration in the Netherlands, 55% originates from sources for which the emissions have not been recorded in the national and European emission inventories. These emissions are primarily caused by natural sources. This portion is referred to as 'non-modelled'. The remaining (modelled) portion of the particulate matter concentration originates entirely from anthropogenic emissions (see the text box *Composition of fine particulate matter in model terms*).

Comparisons of measurement results and model values for the period between 1995-2003 show that the non-modelled fraction differs in magnitude from year to year. However, a significant declining trend was not found (Matthijssen, 2005). This suggests that the contribution of anthropogenic components or related particulate matter fractions in the non-modelled portion is relatively small and more or less constant in time.

The anthropogenic portion of particulate matter in the Netherlands is composed for one-third of primary particulate matter, and the rest is secondary particulate matter (*Figure 5.2*). Half of the primary particulate matter originates from sources in the Netherlands, the other half originates from abroad. Two-thirds of the secondary portion, which is formed in the atmosphere and remains longer in the atmosphere, origi-

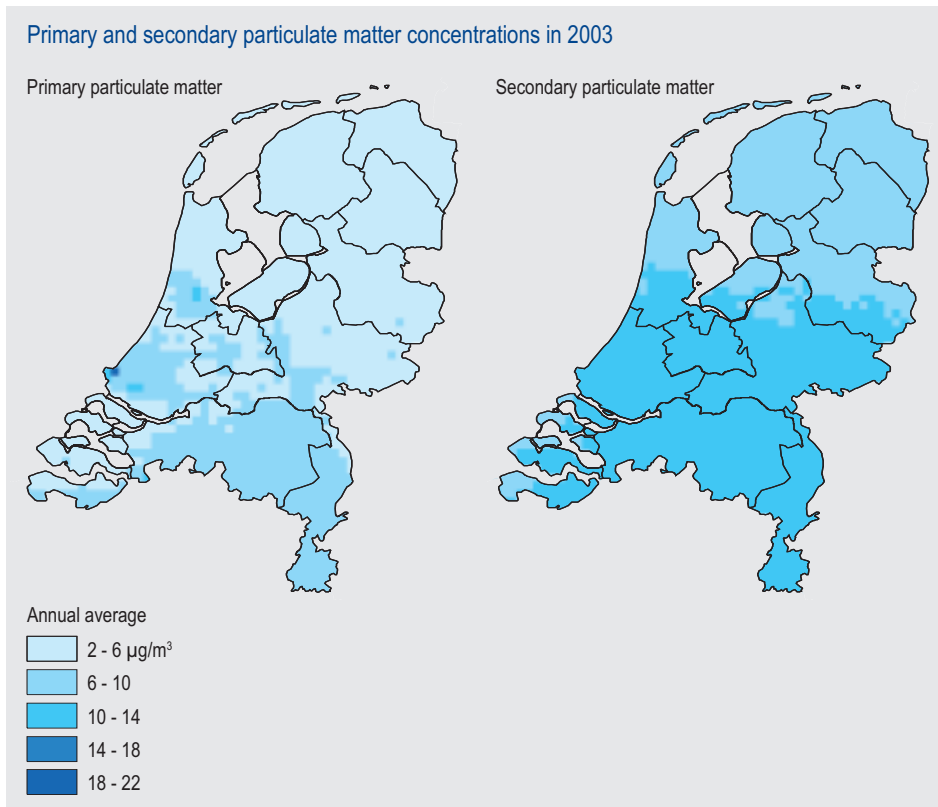


Figure 5.2 Calculated concentrations of primary and secondary particulate matter in 2003, resulting from anthropogenic emissions in the Netherlands and abroad. The concentration field of primary particulate matter has large local variations, primarily in the Western urban agglomerations, while the concentration field of secondary particulate matter is relatively even across the entire country.

nates from abroad. Taken all together, at least 15% of the total background concentration in the Netherlands ultimately originates from anthropogenic sources in the Netherlands. For that matter, the Netherlands is simultaneously a net exporter of particulate matter (Folkert *et al.*, 2005). This means that the quantity of particulate matter of Dutch origin that is transported abroad is greater than the quantity of particulate matter that is transported from abroad to the Netherlands. The Dutch export of particulate matter is three times as large as the import.

The contribution of sources in the Netherlands and abroad to the background concentration in the country as a whole and in the two agglomerations, Rotterdam/Dordrecht and Heerlen/Kerkrade, is illustrated in detail in Figure 5.3. Whereas road transport is the biggest source in the Netherlands, industry, the energy sector and refineries taken together form the largest contribution from sources abroad. Logically, the contribution from abroad is relatively greater in Heerlen/Kerkrade than in Rotterdam/Dordrecht.

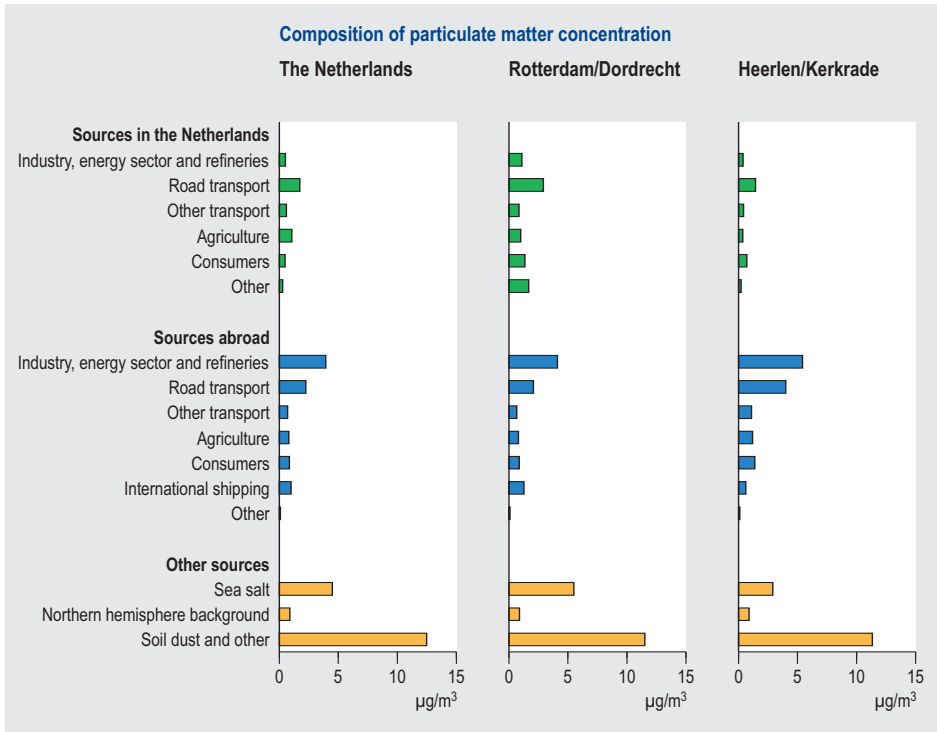


Figure 5.3 Contributions of sources in the Netherlands and abroad to the annual average concentration of particulate matter on average for the Netherlands and for two agglomerations. ‘Soil dust and other’ in the category ‘other sources’ is the many-year average of the non-modelled portion of particulate matter comprising biological material, water, and the contribution of non-modelled or incorrectly-modelled sources. Consequently, this can also partially include anthropogenic sources. For an extensive explanation, see Chapter 4, ‘How much particulate matter do we measure?’ and the text box ‘Chemical composition of particulate matter in the Netherlands’. See also the text box ‘Composition of particulate matter in model terms’.

## Local concentration increases due to traffic in the Netherlands

In urban areas in the Netherlands, the anthropogenic contribution is greater than the national average of 15%. This extra anthropogenic contribution in urban areas is primarily caused by traffic. If the local traffic contribution is added to the total background, the total Dutch contribution to particulate matter at the street level rises to 45%. At specific locations near strong particulate sources, the total Dutch contribution may even rise above 50%.

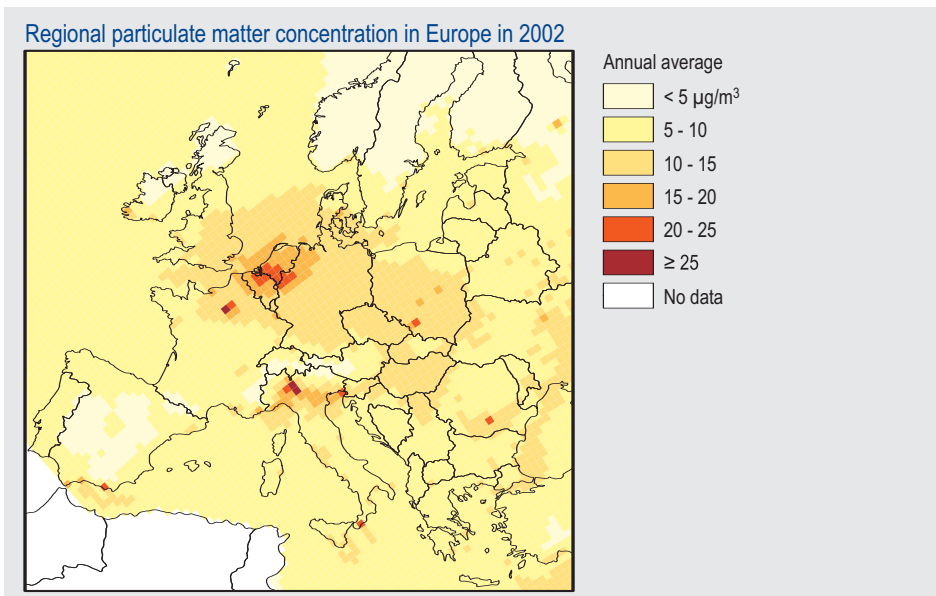
Based on calculations with the CAR model (see the text box *Methodology for calculating particulate matter concentrations*) involving more than one thousand streets in the inner cities of Amsterdam and Utrecht, a picture was obtained of the local traffic contribution to the particulate matter concentration in those streets for the year 2002. In 5% of these streets, local traffic contributes more than 12  $\mu\text{g}/\text{m}^3$  to the particulate matter concentration, in 45% of the streets this contribution is 3-12  $\mu\text{g}/\text{m}^3$  and in 50%

of the streets it is  $0\text{--}3 \mu\text{g}/\text{m}^3$ . If the traffic volume remains the same, in 2010 the total traffic contribution will have declined between 35% and 50%, because the average automobile will emit less particulate matter at that time. The number of streets with a relatively small local traffic contribution ( $0\text{--}3 \mu\text{g}/\text{m}^3$ ) will then rise, according to estimates, from 50% to 75%.

By calculating the background concentration (Steps 1 and 2 in the text box *Methodology for calculating particulate matter concentrations*) separately from the local contribution (Step 3), the contribution from traffic is included twice in the calculation. In the case of inner city streets, this effect can be disregarded. However, if the traffic contribution from busy roads that is calculated with the CAR model is added to the background concentration, the local traffic contribution is overestimated by 8% (Velders *et al.*, 2005).

## Background concentration in Europe

Calculations with the EMEP model used in the European context with a calculation resolution of  $50 \times 50 \text{ km}$  have shown that the background concentrations of particulate matter in the Netherlands are among the highest in Europe, and are similar to the concentrations in parts of Belgium, the Ruhr district of Germany, North Italy and the region around Paris (*Figure 5.4*). The particulate matter concentrations for the



*Figure 5.4 Annual average background concentration of particulate matter in 2002 modelled at ground level in the EU; it is based on anthropogenic emissions and sea salt, Source: EMEP, 2004. The concentration field was calculated using the EMEP model. The results of the EMEP model have not been calibrated to measurements. Therefore the results for all of Europe are too low in comparison with measurements. The relatively high, calculated concentrations in the Netherlands have not been confirmed by the OPS model.*



## Methodology for calculating particulate matter concentrations

The methodology for calculating the particulate matter concentration at any arbitrarily-chosen location in the Netherlands can be divided into three steps. These are described briefly below.

### Step 1. Calculating the background concentration

This concerns the calculation of the background concentration (the regional concentration and the urban concentration) with the OPS calculation model (Van Jaarsveld, 2005). In this model, source contributions in all of Europe are included. Primary and secondary (sulphate, nitrate, ammonium) fractions are calculated separately and then added together to obtain the total calculated particulate matter concentration. The calculation resolution is 5×5 km. The inputs for the model include data about emissions, such as the strength of emissions and spatial and temporal distribution of the sources, both for the Netherlands and for other European countries. Meteorological data are also required. For calculations involving years from the past, the emission data for the Netherlands from the Emission Inventory (MB, 2005; MNC, 2005) and meteorological data for the relevant year are used. For calculations involving future years, the future emissions are estimated based on assumptions about developments of economic activities and emission factors, along with many-year average meteorological input (1990-1999). In the future prognoses, the effect of established national and international policy is taken into account.

### Step 2. Calibration to measurements

This is the calibration of the background concentrations based on measurements from the Dutch Emission Inventory. The results after Step 2 are known as Generic Concentration Maps of the Netherlands and are made available by the Netherlands Environmental Assessment Agency. Calibration is required because the calculated concentrations are about 50% lower than the measured concentrations. This is because the emissions that are used as inputs for the model calculations only concern the known (registered) anthropogenic emissions. Natural sources are not included in the calculations, partly due to a lack of process expertise, but especially due to a lack of reliable emission data. However, measurements provide a total amount of particulate matter comprising particles from both natural

and anthropogenic sources. When the Generic Concentration Maps of the Netherlands are made, this difference is corrected (calibrated) by interpolating differences between the regional background stations across the entire country, and adding this result to the values calculated with the model. For that matter, a similar difference between measurements and calculations is found for all models that are used internationally (EU, 1997; EU, 2004). For the Generic Concentration Maps of the Netherlands, the resolution is increased afterwards from 5×5 km to 1×1 km with a spline-interpolation method (Velders *et al.*, 2005) to better express spatial gradients near cities and point sources for local air quality calculations.

### Step 3. Calculating local contributions

Step 3 concerns the calculation of the contribution of local sources on top of the background concentration from the Generic Concentration Map of the Netherlands, such as a street in an urban environment. The CAR model calculates the particulate matter concentration near roads (Teeuwisse, 2005) and is used to calculate where the limit value for the 24-hour average and the annual average has been exceeded for specific streets. To determine where the limit value for the 24-hour average has been exceeded, the CAR model uses an empirical linear relationship between the annual average particulate matter concentrations and the number of days with a 24-hour average particulate matter concentration above 50  $\mu\text{g}/\text{m}^3$  (Figure 4.3). The linear relationship is based on the measurement results from the Dutch National Air Quality Monitoring Network. This relationship is robust because it has an explained variance of more than 90%. However, this relationship is somewhat different for each type of monitoring station. Moreover, it is probable that the relationship changes through the years. Based on this relationship, the CAR model converts the annual average concentration into a number of days that the limit value for the 24-hour average is exceeded. This relationship shows that the EU limit value, which allows for a maximum of 35 days per year above 50  $\mu\text{g}/\text{m}^3$ , corresponds with an annual average particulate matter concentration of about 31  $\mu\text{g}/\text{m}^3$ . The limit value for the 24-hour average is consequently more stringent than the limit value for the annual average concentration.

Netherlands calculated by the EMEP model are 5  $\mu\text{g}/\text{m}^3$  higher than the national calculation results (and the results of the measurements). However, the Netherlands still emerges as a region with a high background concentration, which is certainly confirmed by the measurements. At the scale level of cities and individual roads, however, this situation is different, and in many European countries there are concentrations at levels that are equal to or higher than those in the Netherlands (*Table 2.1, Table 2.2, Figure 2.2*).

## Uncertainties in the methodology

The measurement results of particulate matter at regional stations are used to calibrate the OPS model calculations. Consequently, they cannot be used once again to independently validate the concentration levels. However, the uncertainty in the calibrated model results can be derived based on all uncertainties (including estimated uncertainties) in the model processes, the input data and the measurements. This results in a total estimated uncertainty in the calibrated, annual average background concentration in the Netherlands of about 40% (95% confidence interval). This concerns the uncertainty for each section on the model grid of  $5 \times 5$  km. Due to the calibration process, the particulate matter concentration that is found is influenced by systematic deviations in the measurements.

Measurements of the natural particulate matter fractions will lead in the future to more reliable estimates of particulate matter concentrations. For prognoses purposes, the uncertainty of particulate matter concentrations is higher than the 40% referred to above, because the expected non-modelled portion is based on an average estimate of a series of years from the past. This series from the non-modelled portion shows variations from year to year, which results in extra uncertainty. The total uncertainty in prognoses is estimated to be at least 50% (95% confidence interval) for the annual average particulate matter concentrations at regional stations. The uncertainties in local street concentrations, such as those calculated with the CAR model, can be estimated by means of comparison with street measurements. This shows that the results of the calculations lie within 30% of the measurements results, but that the results of the calculations are on average about 15% higher than the measurements results.

## Comparison with measurements results and other models

Although the methodology cannot be validated with measurement results of particulate matter, specific fractions of particulate matter, primarily anthropogenic ones such as the sulphate, nitrate and ammonium aerosol, can be validated with measurements. Comparisons with measurements of these components show that both the absolute levels and the trend in the concentrations of these components are in accordance on a yearly average (*Table 5.1; Van Jaarsveld, 2005*).

Table 5.1 Concentrations of secondary particulate matter and primary PM<sub>2.5</sub> as calculated with the EMEP and OPS models for 2002, based on the same emissions. The measurement data were acquired from the Dutch National Air Quality Monitoring Network.

Component	Measurements	EMEP model	OPS model
	Average of 7 stations	The Netherlands (50×50 km)	The Netherlands (5×5 km)
	$\mu\text{g}/\text{m}^3$		
Sulphate aerosol	2.8	3.0	1.8
Nitrate aerosol	3.7	7.3	4.4
Ammonium aerosol	1.9	3.3	1.3
<b>Total secondary particulate matter</b>	<b>8.4</b>	<b>13.7</b>	<b>7.5</b>
Primary PM <sub>2.5</sub>	-	3.9	3.7

The OPS model is the most important policy instrument in the Netherlands for calculating the background concentrations of particulate matter. In a European context, the RAINS model is the most widely used. In contrast to the methodology used in the Netherlands, the underlying EMEP model, and therefore the RAINS model itself, is not calibrated to measurements. Comparisons of the model results show that the modelled primary fractions (in this case PM<sub>2.5</sub>) are in accordance (Table 5.1). In contrast, the secondary fractions modelled by the OPS and the EMEP model differ significantly. The EMEP model calculates values for this fraction that are almost twice as high (5  $\mu\text{g}/\text{m}^3$ ) as calculations from the OPS model. For the Netherlands, it is especially nitrate and ammonium aerosols that are calculated at a higher level by the EMEP model than those that are measured or modelled with the OPS model. As a result, the EMEP model also estimates the particulate matter concentration in the Netherlands higher than in other European countries. In other parts of Europe, a comparable difference with the EMEP model is not found, or at least the differences are not so large.

The comparison with the measurement results is indicative, because the measurements concern an average from a limited number of stations. In contrast, the model calculations are averages for the entire country. An international review has taken place concerning the EMEP model results. Regarding the sulphate and nitrate aerosol, a comparison with the OPS model results has been made that supports the differences reported here (Velders *et al.*, 2003).

### Composition of particulate matter in model terms

The chemical composition of particulate matter is discussed in Chapter 4, *How much particulate matter do we measure?* Models take a different approach: they primarily look to the origin, for example an economic activity or natural source. The OPS model calculates particulate matter concentrations based on registered anthropogenic emissions. The non-modelled portion is – by definition – the difference between the measured and the calculated concentration.

#### Modelled portion

The OPS model calculates the particulate matter concentrations based on anthropogenic emissions. During this process a distinction is made between primary and secondary fractions. The primary fraction is composed of particles that are emitted into the air directly as a result of human activities. The secondary fraction is composed of particles that are formed in the atmosphere after chemical reactions take place in the air. During this process, gases and the particles that are already present both play a role. Ammonia ( $\text{NH}_3$ ), nitrogen oxides ( $\text{NO}_x$ ) and sulphur dioxide ( $\text{SO}_2$ ) are included in the OPS model.

- **Primary sources in the Netherlands.** The most important anthropogenic emissions originate from transport, industry and agriculture. Around 8% of the total concentration results from primary emissions in the Netherlands.
- **Primary emissions from abroad.** The most important sources are industry, transport and consumers. Approximately 10% of the total concentration in the Netherlands results from primary emissions elsewhere in Europe.
- **Secondary emissions in the Netherlands.** The most important sources are transport and agriculture. About 7% of the total concentration in the Netherlands results from secondary emissions from the Netherlands.

- **Secondary emissions from abroad.** The most important sources are transport, industry and agriculture. About 20% of the total concentration in the Netherlands results from primary emissions from elsewhere in Europe.

The above emissions can theoretically be influenced by national and international policy.

#### Non-modelled portion

The non-modelled portion primarily comprises components of natural origin. These emissions can generally not be influenced by policy.

- **Sea salt** In coastal areas, the sea is an important natural source of particulate matter in the form of sea-salt particles. About 14% of the total particulate matter concentration in the Netherlands is composed of sea salt.
- **Soil dust** Soil dust is partially of natural origin. Emission of soil dust can be significantly increased by human activities (agriculture and traffic) and can therefore be classified as anthropogenic. About 12% of the total particulate matter concentration in the Netherlands is composed of soil dust.
- **Northern hemispherical background** This concerns contributions from sources outside Europe, partly of natural origin and partly of anthropogenic origin. The northern hemispherical background contributes about 3% to the total particulate matter concentration in the Netherlands.
- **Other** This is the final item between measurements and calculations. It is composed of biological matter (such as the decomposition products of organic matter and bacteria) water and the contribution of non-modelled or possibly incorrectly-modelled sources and systematic measurement errors. This concerns 27% of the total particulate matter concentration in the Netherlands.

## 6. What are the health effects?

- Public health studies indicate that several thousand people in the Netherlands die prematurely each year in relation to short-term exposure to particulate matter. The temporal magnitude of the premature mortality is probably rather small: several days to several months. Comparable results have been found not only in the Netherlands, but also everywhere in the world, and they are fairly robust.
- If certain American studies about long-term exposure are applied to the Netherlands, it is possible that ten thousand to several tens of thousands of people could die approximately ten years prematurely. However, these results are very uncertain.
- Detrimental health effects have been found for both  $PM_{10}$  and for  $PM_{2.5}$ . It is not yet understood which chemical components of particulate matter cause these health effects. It is virtually certain that sea salt is not hazardous. This is probably true for the sulphate and nitrate fractions in particulate matter as well. However, the soot from combustion processes probably does play a role in the health effects.
- In public health studies, no lower threshold has been found for the health effects of particulate matter. This means that for the time being, there is no outdoor air concentration below which no detrimental health effects can be found.

### *Overview of this chapter*

This chapter addresses the health effects of particulate matter, where the effects of short-term and long-term exposure are discussed separately. The standpoints of the World Health Organisation and of the European Commission are discussed. The uncertainties in the health effects are large, and are therefore given attention separately.

### **Detrimental health effects**

Epidemiological research has shown that exposure to particulate matter in outside air is associated with a wide range of health effects (Brunekreef and Holgate, 2002). These health effects are expressed in phenomena such as premature death, increased hospital admissions for heart and respiratory illness, respiratory symptoms and functional disturbances.

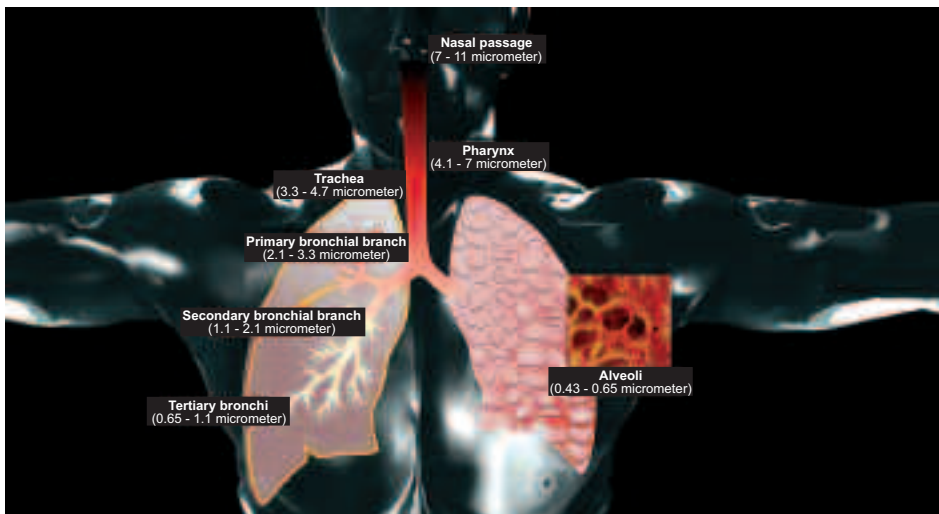
In epidemiological studies, no lower threshold has been found for the health effects of particulate matter. Therefore it is assumed that there is no concentration in the outside air below which no detrimental health effects occur. Health effects have also been observed at concentration levels of particulate matter below the current limit values. The studies on health effects of particulate matter do not indicate which people will suffer damage to their health. However, it can be assumed that the health risk is greater if there is a higher exposure concentration and a greater susceptibility. Susceptible groups include the elderly and individuals with heart disease, circulatory disease or lung disease.

## Fractions of particulate matter that are relevant to health

The various size fractions and the chemical composition of particulate matter components are important to its health effects. During inhalation, particulate matter is deposited at various locations in the airways and lungs. The following rule usually applies: the smaller the particles, the more deeply they penetrate into the airways and lungs (*Figure 6.1*). It is generally assumed that  $PM_{2.5}$  has the most relevance to health, but the coarser component of particulate matter, with a diameter between 2.5 and 10 micrometers, can certainly not be disregarded.

It is not yet well understood which chemical components of particulate matter are most relevant to health. However, it does seem to be clear that sea-salt aerosol and the secondary inorganic fractions, such as sulphate and nitrate aerosol, have little importance to the direct health effects of particulate matter (Schlesinger and Cassee, 2003). Regarding soil dust, windblown or otherwise, there is still insufficient information to determine whether, and to what extent, this fraction is relevant to health.

Various combustion sources contribute to the emission of primary, anthropogenic particulate matter fractions, including soot. Examples of such combustion sources include traffic, shipping, industry, energy generation and domestic heating. It is not yet possible to quantify the health effects of these sources. However, there are indications that emissions from traffic play a role in the health effects that are related to short-term and long-term exposure. This certainly applies to urban areas and to busy traffic situations. Policy that focuses on reducing the emission of the primary fractions of particulate matter could therefore be beneficial in terms of public health.



*Figure 6.1 Schematic diagram of the human respiratory system. The particle sizes in parentheses are only indicative, but they do illustrate that the smallest particles can penetrate the deepest into the lungs. Source: Trouw. © Trouw, 2005.*

In addition, a possible role for semi-volatile components has been taken into account regarding the health effects. The same applies to fractions of biological origin such as pollen. For people who are sensitive to such fractions, these biological components can cause significant effects. In this context, there are indications of a combined effect of pollen and traffic emissions.

## Assessment by the World Health Organisation and the European Commission

In 1987 and 2000, the World Health Organisation (WHO) published summaries of the health effects of air pollution. Recently, the WHO conducted another similar evaluation that was the basis for the Clean Air for Europe (CAFE) programme of the European Commission (WHO, 2004a, 2005). Once again it was ascertained that no lower threshold value for health effects can be observed for particulate matter.

For this reason, the WHO did not establish an air quality guideline for particulate matter. However, a concentration/effect relationship has been established for the health effects that are related to long-term exposure to particulate matter. As part of this concentration/effect relationship, the WHO proposed  $PM_{2.5}$  as a new indicator; it is thought that it is primarily this fraction that is important to health. Due to the lack of European data from long-term cohort studies, a risk factor from one of the major American studies (Pope *et al.*, 2002) was used and was declared to be applicable to Europe. The effect data linked to this study therefore formed the basis, in part, of the current CAFE calculations.

For purposes of comparison, the American Environmental Protection Agency (US EPA) established a limit value for  $PM_{2.5}$  a number of years ago, applying a limit value of  $15 \mu\text{g}/\text{m}^3$  for the annual average and a limit value of  $65 \mu\text{g}/\text{m}^3$  for the 24-hour average. At the present time, the US EPA is revising the limit values for air quality, also with the option of establishing a new indicator; this would be a limit value for 24-hour average concentrations of  $PM_{10-2.5}$ .

Due to the lack of sufficient numbers of studies, it turned out to be impossible to establish a generally applicable concentration/effect relationship for short-term exposure to  $PM_{2.5}$  based on European effect data (WHO, 2004b). The calculations used in CAFE for short-term exposure to particulate matter are therefore based on risk figures from various studies. As part of the CAFE strategy, there will probably not be a proposal to change the current limit values for  $PM_{10}$ . However, there are proposals in the European Commission to add  $PM_{2.5}$  as a new, extra indicator, in addition to  $PM_{10}$ , and to establish a corresponding limit value.

**Possible mechanisms for health effects**

The biological mechanisms with which particulate matter can cause hazardous effects are not well understood. Nevertheless, particulate matter does not appear to cause death or illness in a healthy person. However, it does appear to exacerbate existing illnesses, especially serious respiratory conditions such as asthma, as well as heart and circulatory disease. Consequently, people with these conditions appear to have the greatest health risk. The toxicological research into particulate matter has presented some supportive data for the health effects, but can not yet provide clarity about the causality in the observed exposure/effect relationships; neither can it indicate which components from the complex mixture of particulate matter are the most responsible for the health effects.

During this process there have been several ideas presented about the origin of the haz-

ardous effects. During inhalation, particulate matter enters the upper and lower airways and the lungs. At these locations it can cause inflammation reactions and can hamper the intake of oxygen. With people who already have weak lungs due to other causes, this can ultimately be fatal. These inflammation reactions, and the free radicals that are released in the process, can also damage the heart function and therefore be hazardous to heart patients. Particulate matter may also affect the coagulation balance in the blood, causing blood to become more viscous and leading to an increased risk of a heart attack. In addition, neurological effects of particulate matter have also been found where, for example, the heart function or heart muscle function can be negatively affected. It is assumed that these types of effects also contribute to a process of accelerated ageing.

**Health effects related to short-term exposure**

Associated with short-term exposure to particulate matter, it is estimated that 2300-3500 people die prematurely each year in the Netherlands, with 3000 as an average (MNP, 2005). These estimates are in accordance with previous reports. The reduction in life expectancy is probably small, ranging from several days to several months. Research from around the world, and from the Netherlands as well, has indicated the existence of such relationships.

**Health effects related to long-term exposure**

There is concern about a possibly greater effect on health that is related to long-term exposure to particulate matter. This refers to the air pollution level to which people are exposed for multiple years or for their entire lives. Reliable data from the Netherlands and the rest of Europe to estimate this risk are still lacking. When such estimates are made based on the results of two large-scale American studies (Dockery *et al.*, 1993; Pope *et al.*, 1995, Pope *et al.*, 2002), the magnitude and severity of these effects appear to be greater than the effects that are associated with short-term exposure.

In their study, Knol and Staatsen (2005), estimated that the magnitude of these effects in the Netherlands would be between 12,000 and 24,000 premature deaths per year with 18,000 as an average, at an annual average particulate matter concentration of 35  $\mu\text{g}/\text{m}^3$ . This range in premature mortality is the result of the statistical uncertainty of the two underlying individual studies and their mutual deviation. These statistical margins should be seen as a global indication.



Moreover, due to additional elements of uncertainty, the total uncertainty for the application of the American studies to the situation in the Netherlands is significantly larger. The magnitude of these recent estimates is reasonably in agreement with previous reports on this topic (Buringh and Opperhuizen, 2002a, 2002b). The estimated reduction in life expectancy that occurs is tentatively estimated at ten years. However, this estimate is also uncertain. In the *Milieubalans 2005*, this risk estimate was expressed in DALYs per 1000 population (MNP, 2005; see below).

A study was recently launched in the Netherlands concerning the health effects that are related to chronic exposure to air pollution (Hoek *et al.*, 2002). In several years, more information will therefore be available about the possible effects in the Netherlands. This could then be used in part as the basis for an up-to-date risk assessment.

## Uncertainties in the risk assessment

The current level of knowledge about the health effects of long-term exposure to particulate matter is low and the uncertainties in the above assessments are therefore large. The level of knowledge concerning the effects of short-term exposure is higher, and the uncertainties are smaller. The small number of long-term effect studies about particulate matter and their sometimes conflicting results are the most important causes of this low level of knowledge.

For example, an American study of war veterans did not show any association, while a study of Seventh Day Adventists (a group which is expected to have good general health) had results that differed for men and women (see also Knol and Staatsen, 2005). However, the American studies used for the assessment in the Netherlands are consistent, and one of the studies also showed the same results when the study continued for a longer period. These studies were conducted with large groups of people and are considered to be robust.

However, the question remains of whether these data are equally applicable to the situation in the Netherlands. The WHO and the European Commission have proposed to use this American data as a basis for European policy. In view of the uncertainties, there is also a chance that the actual health risks of particulate matter will be assessed as too low.

The uncertainties in the risk assessment primarily concern the following issues:

- the question of whether or not the observed statistical correlation from the epidemiological research indeed has a cause-and-effect relationship, if the correct particulate matter indicator was used and if there has been sufficient correction for other distorting variables;
- the question of whether research data from other countries can be applied to the exposure situation in the Netherlands due to differences in the population, the composition of the particulate matter and the other aspects of air quality;
- the assessment of the magnitude and duration of the various effects;

- the statistical uncertainties in the assessment of the risk factors;
- the question of whether the relationships found are indeed linear;
- the decision of whether or not to use a threshold value and revert to a hypothetical concentration without any particulate matter in the outside air.

## The illness burden in the population: DALY

Besides being expressed in numbers of people, health effects in the population are currently also expressed in a new health indicator, the DALY (Disability Adjusted Life Years). Briefly summarised, one DALY unit means that one human being dies one year earlier. This indicator takes account of the magnitude, severity and duration of the effects. As a result, it establishes a kind of universal health indicator for the total of mortality and illness in the population.

The health effects that are related to long-term exposure to particulate matter can also be expressed in DALYs. The premature mortality by itself, assuming a level of 18,000 premature deaths, leads to 180,000 DALYs for the Dutch population. However, the uncertainty in this estimate is extremely large. Knol and Staatsen (2005) arrived at an estimate of this long-term effect of particulate matter that varied from several percent to perhaps more than 15% of the calculated, total illness burden in the population.

A similar calculation for other effects of chronic exposure to particulate matter is not possible at this time because there are too few data available to make such a calculation. The magnitude of the total illness burden of the population is, when all health effects of particulate matter in the long term are taken into account, therefore still unknown.

## Improved public health through policy

A number of studies have been published about the reduction of detrimental health effects due to the influence of policy and by means of specific emission interventions, including interventions involving particulate matter. Many of these studies concern situations with relatively high concentrations. Health benefits such as lower mortality, reduced illness and less reduction in life expectancy are sometimes predicted by models when there is a reduction in the total mass of particulate matter. In view of our current state of knowledge, however, such an assessment of the possible health benefits is very uncertain.

The health benefits can be overestimated when the mass reduction in particulate matter is primarily the result of a reduction of fractions that have little relevance to health, such as sulphate or nitrate aerosol. The health benefits can be underestimated if there is a larger mass reduction of a fraction that has a greater effect on health, such as the soot fraction, compared to the mass reduction of particulate matter as a whole. Insight into how policy in the area of air quality affects such particulate matter fractions, and the health benefits that are linked to such policy, is only possible when the causality is better understood.

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## Information on the Internet

The Internet is a good and extensive source of information on particulate matter. A number of links are listed below where you can find more information about specific aspects of the particulate matter problem.

### Policy in the Netherlands

- Ministry of Housing, Spatial Planning and the Environment; Air Quality Dossier

### European policy

- European Union, air quality
- The Clean Air For Europe (CAFE) programme

### Institutes and organisations active in the area of the environment

- Environmental Protection Agency, U.S.A.
- European Environment Agency, Copenhagen
- European Topic Centre on Air and Climate Change, Bilthoven
- The Interregional Cell for the Environment, Brussels
- The Netherlands Environmental Assessment Agency
- The National Institute for Public Health and the Environment/Public Health
- The National Institute for Public Health and the Environment/Environment and Safety
- The Federal Environmental Agency, Berlin
- Flemish Environment Agency, Antwerp

### Monitoring results in the Netherlands

- Dutch National Air Quality Monitoring Network
- DCMR Environmental Protection Agency Rijnmond

### Monitoring results in other countries

- Belgium, Flanders, Flemish Environment Agency
- Germany, Nordrhein-Westfalen, Landesumweltamt Nordrhein-Westfalen
- Europe, AIRBASE, the air quality database of the European Topic Centre on Air and Climate Change

### Environmental Quality in the Netherlands

- Environmental Data Compendium

The dossier on Transboundary Air Pollution on the website of the Netherlands Environmental Assessment Agency provides a summary of the hyperlinks of the institutes and organisations that are listed above.